FIELD COMPARISON OF MEASUREMENT METHODS FOR NITRIC ACID

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Final Report

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Abstract

Eighteen instruments for measuring atmospheric concentrations of nitric acid were compared in an eight day field study at Pomona College, situated in the eastern portion of the Los Angeles Basin, in September 1985. The study design included collocated and separated duplicate samplers, and the analysis by each laboratory of a set of quality assurance filters, so that the experimental variability could be distinguished from differences due to measurement methods.

For all sampling periods, the values for nitric acid concentrations reported by the different instruments vary by as much as a factor of four. The differences among meausurement techniques increased with nitric acid loading, corresponding to a coefficient of variation of 40%. In contrast, samplers of the same design operated by the same group show variability of 11% to 27%.

Overall, the highest reportedconcentrations are from the filter packs, lower concentrations are given by the annular denuders and tunable diode laser absorption spectrometers. When the nitric acid concentrations were high enough to be detected by the FTIR, the FTIR values are close to those obtained by the denuder difference method, and to the mean value from the other sampler methods.

In the absence of a reference standard for the entire study, measurement methods are compared to the average of four denuder difference method samplers (DDM). Filter pack samplers are higher than the DDM for both daytime and nighttime sampling. Two different filter packs using Teflon prefilters are higher than the DDM by factors of 1.25 and 1.4. The results from the three annular denuders do not agree; the ratios of means to the DDM value are 1.0, 0.8 and 0.6. For the transition flow reactor method and for two dichotomous samplers operated as denuder difference samplers, the ratio of means to the DDM are 1.09 and 0.93 respectively. The tunable diode laser absorption spectrometers give lower daytime and higher nighttime readings compared to the DDM, especially during the last three days of the study. Averaged over the entire measurement period, the daytime ratio of TDLAS to DDM is 0.8 and the nighttime ratio is 1.7.

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I. SUMMARY AND CONCLUSIONS

The Nitrogen Species Methods Comparison Study was a field comparison of methods for measuring atmospheric concentrations of nitric acid and other nitrogenous species. Investigators from 20 different research groups participated in the study, which was held at Pomona College in Claremont, CA, located in the eastern portion of the Los Angeles Basin. Instruments were operated side-by-side for an eight day period, from September 11 to September 19, 1985.

The principal objective of the study was to evaluate routine measurement methods for nitric acid which could be used in the Southern California Air Quality Study (SCAQS). Continuous methods for nitric acid included Fourier transform infrared spectroscopy (FTIR, Tuazon *et al.* 1981), tunable diode laser absorption spectroscopy (TDLAS, Schiff *et al.* 1983), and the tungstic acid technique (TAT, Braman *et al.,* 1982). Time integrated techniques included filter packs (FP), the denuder difference method (DDM, Shaw *et al.* 1982 and Appel *et al.* 1981), the annular denuder method (ADM, Possanzini *et al.* 1983), and the transition flow reactor (TFR, Knapp *et al.* 1986).

Other nitrogen species measured during this study included total and fine particle nitrate, ammonia, particulate ammonium ion, nitrous acid, nitrogen dioxide, peroxyacetyl nitrate (PAN), oxides of nitrogen (NO_x) and the nitrate radical. Routine air quality and meteorological parameters such as ozone, temperature and relative humidity were monitored. The study design included collocated and separated duplicate samplers, and the analysis by each laboratory of a set of quality assurance filters, so that experimental variability could be distinguished from differences due to measurement methods.

In this report we present the results for nitric acid. Additionally, we describe the measurement methods and the protocol followed for the data collection, and we list all of the data in tabular format. Although the data presented permit comparisons for many nitrogenous species, the data analysis addresses nitric acid data only.

Results

Principal results with regard to the nitric acid measurement methods are, as follow:

(1) There are statistically significant, systematic differences in the nitric acid concentrations obtained by the different measurement techniques and sampler configurations employed in the Nitrogen Species Methods Comparison Study. For each sampling period in the study, reported nitric acid concentrations vary among samplers by as much as a factor of four. For the period with the highest nitric acid values reported concentrations range from 191 neq/m^3 to 800 neq/m^3. The standard deviation among all reported values for nitric acid increases linearly with the nitric acid loadings, corresponding to a consistent coefficient of variation of 40%. This variation is much larger than for analysis of the filters upon which known amounts of nitrate had been deposited (better than 11% accuracy for most groups), or for replicate samplers operated by the same group (12% to 27% variability).

(2) Overall, the highest concentrations are from the filter packs, lower concentrations are given by the annular denuders and tunable diode laser absorption spectrometers. Values from the denuder difference method and the transition flow reactors are close to the mean of the methods. When the nitric acid concentrations were high enough to be detected by the FTIR, the FTIR values are nearest those obtained by the denuder difference method; however, values from each method are within the reported uncertainty. The mean FTIR value for the high nitric acid sampling periods is within 3% of the mean of the other methods.

(3) The filter pack method gives higher results for both daytime and nighttime sampling. Differences are also seen among the filter packs operated by different groups. Filter pack sampler #GF3 is higher than the DDM by a factor of 1.25 whereas the filter pack #CF1 is higher by 1.4.

(4) In some cases differences exist in the implementation of the measurement method by different groups, as well as differences between types of measurements. The three annular denuders do not give the same results. Nitric acid concentrations from #EA1 are greater than from #QA1, which are greater than from #IA1 (#EA1 > #QA1 > #IA1). For the two TAT systems, nitric acid concentrations from #AC1 were greater than from #TC1. The six denuder difference samplers, including the dichotomous samplers, reported values which are not statistically different from each other. Similarly, values from the two transition flow reactors are not statistically different, nor do the values from the two TDLAS systems differ from one another.

(5) In the absence of a reference standard for the entire study, measurement methods are compared to the average of four denuder difference method samplers. For the annular denuders, the ratios to the DDM value for #EA1, #QA1 and #IA1 are 1.0, 0.8 and 0.6 respectively. For the transition flow reactor the ratio of means to the DDM is 1.09. For the two dichotomous samplers operated as denuder difference samplers, the ratio of means to the DDM is 0.93 .

(6) For three of the samplers, there were large differences in the relative performance between daytime and nighttime sampling. The TDLAS instruments gave low daytime and high nighttime readings in comparison to the other measurements. This is most marked on the last three days of the study. Ratios of means to the DDM are 0.77 daytime and 1.65 nightime. The TAT system #TC1 also was low during the day and high at night. The opposite diurnal response is seen with filter pack #JF1 which was high during the day and low at night. The FP, ADM, DDM and TFR averages do not exhibit significant diurnal variations with respect to the mean of methods.

Conclusions

Due to the lack of an absolute reference standard for the entire study period, we cannot make a definitive statement as to the most accurate nitric acid measurement method. Furthermore, the choice of measurement method depends upon the length of the sampling period and the expected nitric acid concentrations. With these qualifications, our recommendations with regard to monitoring methods for nitric acid are, as follow:

(1) In studies where large nitric acid concentrations are to be measured over short (4-hour) time periods, the denuder difference method appears to be the most accurate and reliable monitoring technique. It is not precise when nitric acid concentrations are low, below about 25 neq/m^3 (0.6 ppb), depending on analytical sensitivity and the HNO_3 to fine particle nitrate concentration ratio.

(2) In studies where total inorganic nitrate concentrations are to be monitored, the filter packs may are a good choice. They give an upper bound on nitric acid and an accurate measure of the sum of particle nitrate and nitric acid. The filter packs gave the most precise and reproducible results, as judged by replicate samplers operated by the same group.

(3) The transition flow reactor and the dichotomous sampler (operated as a denuder difference sampler) gave similar results for nitric acid as the denuder difference method, and should be considered as a possible monitoring method.

(4) The tungstic acid technique and the annular denuder methods require additional development before they can be employed as routine monitoring methods.

II. OVERVIEW

The Nitrogen Species Methods Comparison Study was a jointly sponsored atmospheric field study conducted in the Los Angeles basin in September 1985. The principal objective was to evaluate, under field conditions, the differences among various techniques used for measuring atmospheric concentrations of nitrogenous species. In this work we present the nitric acid results for 18 different instruments.

Instruments for measuring nitric acid were operated side by side for an eight day period, from September 11 - 19, 1985, on the Pomona College campus in Claremont, CA. Continuous methods for nitric acid included Fourier transform infrared spectroscopy (FTIR, Tuazon *et al.* 1981), tunable diode laser absorption spectroscopy (TDLAS, Schiff *et al.* 1983), and the tungstic acid technique (TAT, Braman *et al.* 1982). Time integrated techniques included filter packs (FP), the denuder difference method, (DDM, Shaw *et al.* 1982, and Appel *et al.* 1981), the annular denuder method (ADM, Possanzini *et al.* 1983), and the transition flow reactor (TFR, Knapp *et al.* 1986).

Other nitrogen species measured during this study included total and fine particle nitrate, ammonia, particulate ammonium ion, nitrous acid, nitrogen dioxide, peroxyacetal nitrate (PAN), oxides of nitrogen (NO_x) and the nitrate radical. Routine air quality and meteorological parameters such as ozone, temperature and relative humidity were monitored. This paper will review results only from the nitric acid data.

Some of these nitric acid methods were also compared in the 1979 Claremont study (Spicer *et al.*, 1982). In that study attention was directed toward artifacts arising from the facile conversion between particulate and gaseous nitrates. In the present study attention is also given to differences between daytime and nighttime measurements, and to short-term *vs.* long-term sampling.

III. EXPERIMENTAL PLAN

Study Design

In addition to the differences among measurement methods, there are two other sources of variability in the field study which required evaluation. First, with so many samplers, it was not possible to use a common sampling manifold. The samplers were sited in a 43 m by 12 m area. Therefore, the influence of sampler siting had to be investigated. Second, the samplers were operated by different groups, each of whom were responsible for their own chemical analyses. Hence variability in chemical analyses needed to be assessed.

The study consisted of four parts:

(1) Interlaboratory comparison of analysis of Teflon® and nylon quality assurance filters containing known amounts of nitrate and sulfate.

(2) Replicate measurements with instruments of the same design and from the same research group, located at different positions on the sampling platform.

(3) Side-by-side sampling with all instruments in the field over an 8-day period.

(4) Simultaneous measurements of meteorological data and of potential interferents such as PAN and nitrous acid.

The analysis of quality assurance filters (No. 1) allows us to assess variability not attributable to the sampling method, while replicate units (No. 2) allow us to assess variability due to instrument siting. The side-by-side sampling (No. 3) includes variability due to measurement method, instrument siting and chemical analysis.

Nitric Acid Measurement Methods

The measurements at the site are summarized in Table 1. Most of the nitric acid measurement methods fall into one of eight categories. Time-integrated

Table 1. Sampler Descriptions
Nitrogen Species Methods Comparison Study, 9/85, Pomona College, Claremont, CA

GROUP	ID	SAMPLER	INLET	HLDR	(L/min)	SPECIES
Filter Packs						
Anlauf/Wiebe	CF1	-T/N/Citric Acid	--	T	40	HNO3, NO3-, NH3, NH4+,SO4--
	CF2	-T/N/Citric Acid	--	T	40	HNO3, NO3-, NH3, NH4+,SO4--
Eatough	EF1	-Imp-Q/N	P&G	T	10	HNO3,NO3-,NH4+,HNO2
Cass/Solomon	GF1	-T/N, -T/Ox/Ox	--	P	3, 4.5	HNO3,NO3-,NH3,NH4+,SO4--
	GF2	-T/N, -T/Ox/Ox	--	P	3, 4.5	HNO3,NO3-,NH3,NH4+,SO4--
	GF3	-T/N, -T/Ox/Ox	--	P	3, 4.5	HNO3,NO3-,NH3,NH4+,SO4--
	GF4	-T/N, -T/Ox/Ox: (network)	--	P	3, 4.5	HNO3,NO3-,NH3,NH4+,SO4--
Grosjean	MF1	-N/N & -T/N	--	P	20	Total nitrate, oxalate
Martin/Mitchell	JF1	-T/N/K2CO3	--	T	4	HNO3, NO3-,SO2,SO4--
	JF2	-T/N/K2CO3	--	T	4	HNO3, NO3-,SO2,SO4--
Womack	NF1	El-T/N/S	T	T	3	HNO3,NO3-,SO4--
	NF2	El-T/N/S (6 days/sample)	T	T	3	HNO3,NO3-,SO4--
	NF3	El-T/N/S (6 days/sample)	T	T	3	HNO3,NO3-,SO4--
	NF4	-T/N/S (6 days/sample)	T	T	3	HNO3,NO3-,SO4--
Denuder Difference Methods						
Appel	AD1	Cy-D(MgO)/N, -Cy-N	T&G	P	20	HNO3, NO3-, SO4--
Cass/Solomon	GD1	Cy-{-D(MgO)/N, -N, -T}	G&T	T	3+3+4.5	HNO3, NO3-,SO4--
	GD2	Cy-{-D(MgO)/N, -N, -T}	G&T	T	3+3+4.5	HNO3, NO3-,SO4--
	GD3	Cy-{-D(MgO)/N, -N, -T}	G&T	T	3+3+4.5	HNO3, NO3-,SO4--
Horrocks	FD1	Cy-D(MgO)/T/N, Cy-T/N	T&G	P	20	HNO3, NO3-, SO4--
John	BD1	Dichot{T/N,T/N}	Al	Al	16.7	} HNO3,
	BD2	Cy-T/N	G&T	P	24	} Coarse and fine NO3-, SO4--,
	BD3	D(Anodized Al)/Dichot{T/N,T/N}	Al	Al	16.7	} NH4+,H+,Na+, Ca+,Cl-
Pierson	KD1	Cy-{-D(MgO)/N, -N} -N	T	SS	10+18,18	HNO3, Coarse and fine NO3-,SO4--
	KD2	Cy-{-D(MgO)/N, -N} -N (1/day)	T	SS	10+18,18	HNO3, Coarse and fine NO3-,SO4--
Annular Denuders						
Sickles/	IA1	Cy-D(Na2CO3,Na2CO3,Citric Acid)/	T	T	15	HNO3,HNO2,NO2,NO3-,SO2,SO4--
Allegrini	IA2	/T/N/D(Citric Acid or KI&NaAsO2)	--	--	--	HNO3,HNO2,NO2,NO3-,SO2,SO4--
Eatough	EA1	Cy-D(Na2CO3,Na2CO3)/Q/N	T	T	20	HNO3,HNO2,NO3-,SO2,S04--,AlkSO4
	EA2	Cy-D(Na2CO3,Na2CO3)/Q/N	T	T	20	HNO3,HNO2,NO3-,SO2,S04--,AlkSO4
Peake	QA1	Cy-D(Na2CO3,Na2CO3,Citric Acid)/T/N	T	T	14	HNO3,HNO2,NO3-,NH3,SO2,SO4--

Table 1. Sampler Descriptions
Nitrogen Species Methods Comparison Study, 9/85, Pomona College, Claremont, CA

GROUP	ID	SAMPLER	INLET	HLDR	(L/min)	SPECIES
Transition Flow Reactors						
Ellestad	HE1	Cy-{-D(N,Nafion)/T/N/Ox/TEA/TEA	T	T	18	HNO3,NO2,NO3-,NH3,NH4+,SO2,SO4--
	HE2	-D(N,Nafion)/T/N/Ox/TEA/TEA}	--	T	18	HNO3,NO2,NO3-,NH3,NH4+,SO2,SO4--
Spicer	DE1	Cy-{-D(N,Nafion)/T/N/Ox/TEA/TEA	T	T	18	HNO3,NO2,NO3-,NH3,NH4+,SO2,SO4--
	DE2	-D(N,Nafion)/T/N/Ox/TEA/TEA}	--	T	18	HNO3,NO2,NO3-,NH3,NH4+,SO2,SO4--
Continuous Methods						
Stedman	SC1	Luminol HNO3	T	T	2	HNO3
Winer/Tuazon	RC1	FTIR (HNO3, NH3)	--	--	--	HNO3,NH3
Anlauf/Wiebe	CC1	T/TDLAS (HNO3, NO2)	9' T	T	5	HNO3,NO2
Mackay	PC1	T/TDLAS (HNO3, HCHO)	9' T	T	5	HNO3,HCHO
Appel	AC1	Tungstic Acid Tubes	3' G	--		HNO3,NH3
Braman	TC1	Tungstic Acid Tubes	3' G	--	0.7	HNO3,HNO2,NO3-
Other Gaseous Species						
Appel	AO1	-D(Ox)	--	--	1	NH3
Appel	AF1	-T/Ox/Ox	P	P	25	NH3, NH4+
Winer/Biermann	RC2	DOAS	--	--	--	HNO2, NO2, NO3'
Spicer/Holdren	DC1	PAN - GC		--		PAN
Grosjean	MF2	-KOH/KOH & -DNPH/DNPH	--	P	20	Acids, phenols, PBzN, carbonyls
Grosjean	MF3	-T/KOH & -T/DNPH	--	P	20	Acids, phenols, PBzN, carbonyls
Mackay	PC2	Luminox Analyzer	T	--	--	NO2
Anlauf/Wiebe	CC2	T/N/NOx Box	T	--	--	NO, NOx(NO+NO2+PAN)
Appel	--	Dasibi 1003	--	--	--	O3
Ellis	--	Gases {O3, SO2, NOx,CO}	--	--	--	O3, NOx, CO
Eatough	--	GC-FPD (Dimethylsulfate)	T	--	0.5	(CH3O)2SO2
Other Aerosol Measurements						
John	BO1	Berner Low Pressure Impactor	AI	AI	30	NO3-, NH4+, SO4--, H+,Na+,Cl-,Ca,Mg
Ellis	OO1	Dichotomous Sampler	AI	AI	17	NO3-,SO4--
Lawson	FO1	HiVol/G	AI	AI	1120	NO3-,SO4--,TSP
	FO2	HiVol/Q	AI	AI	1120	NO3-,SO4--,TSP
	FO3	PM10-HiVol/Q	AI	AI	1120	NO3-,SO4--,PM10

Table 1. Sampler Descriptions
Nitrogen Species Methods Comparison Study, 9/85, Pomona College, Claremont, CA

GROUP	ID	SAMPLER	INLET	HLDR	(L/min)	SPECIES
Meteorological Measurements						
Appel	--	Temperature, relative humidity	--	--	--	Temp, RH
Ellis	--	Temperature, relative humidity, winds, barometric pressure, solar intensity.	--	--	--	met data
Special Experiments						
Eatough	EO3	Im-D(Ox,Ox,Ox,Ox,)/Q/N/Ox	P&G	T	2	NO3-, NH3, NH4+
	EO2	Im-{-D(TA,TA,TA,TA)/Q/N/Ox	P&G	T	2	HNO3,NO3-,NH3,NH4+
	EO1	Im-{D(N,N,N,N,N,N,)/Q/N	P&G	T	2	HNO3,NO3-, AlkSO4, HNO2
	EO5	-D(N)/Q/N}	P&G	T	10	HNO3,NO3-
	EO7	-D(6xNa2CO3)/Q/N	P&G	T	2	HNO3,HNO2,NO3-
Ellestad	HO1	Cy-D(N)/T/D(N)/N (4 days/dy-nt pr)	T	T	1.5	HNO3
Filter Tests	TO	-T/N(old Membrana)	--	P	15	HNO3,NO3-,SO4--
	TL	-T/N(new, low pressure drop, #871)	--	P	15	HNO3,NO3-,SO4--
	TH	-T/N(new, high pressure drop, #4195)	--	P	15	HNO3,NO3-,SO4--
Deposition Measurements						
John	BO2	Plants & Artificial Surfaces	--	--	--	NO3-, SO4--
Pierson	KO1	Dew Collectors	--	--	--	HNO3,NO3-,SO4--
Pierson	KO2	Surrogate surfaces	--	--	--	HNO3,NO3-,SO4--
Atmospheric Mutagens						
Winer/Atkinson	RX1	MegaVol/TIGF & G	Al	Al	17900	Mutagenicity & Nitroarenes
	RX2	PM10-HiVol/TIGF	Al	Al	1120	Mutagenicity & Nitroarenes
		PM10-HiVol/G	Al	Al	1120	Mutagenicity & Nitroarenes
	RX3	Tenax columns	--	--	2	Reactivity of pyrene & fluoranthene

Symbols:

HLDR: Holder	Q: Quartz Filter
T: Teflon	N: Nylon Filter
G: Glass	Ox: Oxalic acid impregnated filter
Al: Aluminum	TA: Tungstic acid
P: Plastic	TEA: Triethanolamine impregnated filter
	TIGF: Teflon impregnated glass fiber filter
	AlkSO4: alkylated sulfates

samples were collected by (1) filter packs, (2) denuder difference samplers, (3) annular denuders and (4) the transition flow reactor. Continuous and semi-continuous methods include (5) tunable diode laser absorption spectroscopy (6) Fourier transform infrared spectroscopy, (7) tungstic acid hollow tube denuder collectors, and (8) a luminol method for nitric acid. Descriptions of these methods are given in Appendix A.

The configuration for each sampler is outlined in Table 1. The method used to obtain a fine particle precut is indicated by "Cy", "Imp" or "El" for cyclone, impactor and elutriator, respectively. The types of filters used are given by "T" for Teflon®, "N" for nylon, "Ox" for oxalic acid impregnated, etc. The MgO denuders are shown as "D(MgO)", the carbonate denuders by "D(Na2CO3)". The research groups are identified by the principal investigator(s). The first letter in the sampler number is the identifier for the research group; the second letter indicates the measurement method.

Sampling Site and Measurement Protocol

Sampling was conducted at the Pomona College campus in Claremont, CA., located in the eastern part of the Los Angeles Basin, as shown in Figure 1. The site was situated in an unused parking lot along the southeastern edge of the campus. The layout of the site is shown in Figure 2. Except for the two diode laser systems and one of the tungstic acid tube systems, all of the instruments were located outside. Most were placed in a line along a 1 m high, 43 m long platform oriented perpendicular to the prevailing afternoon winds. Sampling inlets were positioned 1.5 m above the sampling platform (2.5 m above the ground). The FTIR operated with a 25 m open path located 3 m in front of the platform and 2.5 m above the ground. Thus all sampler inlets and the FTIR basepath were 2.5 m above the ground. This configuration was used to minimize the influence of possible vertical concentration gradients.

During the first two days of the study, all replicate instruments, i.e. identical instruments operated by the same group, were positioned side by side. For the remaining six days of the study, several of the replicate instruments were sited at different positions along the platform. This design permitted evaluation of differences due to sampling location. The location of each of the samplers for these

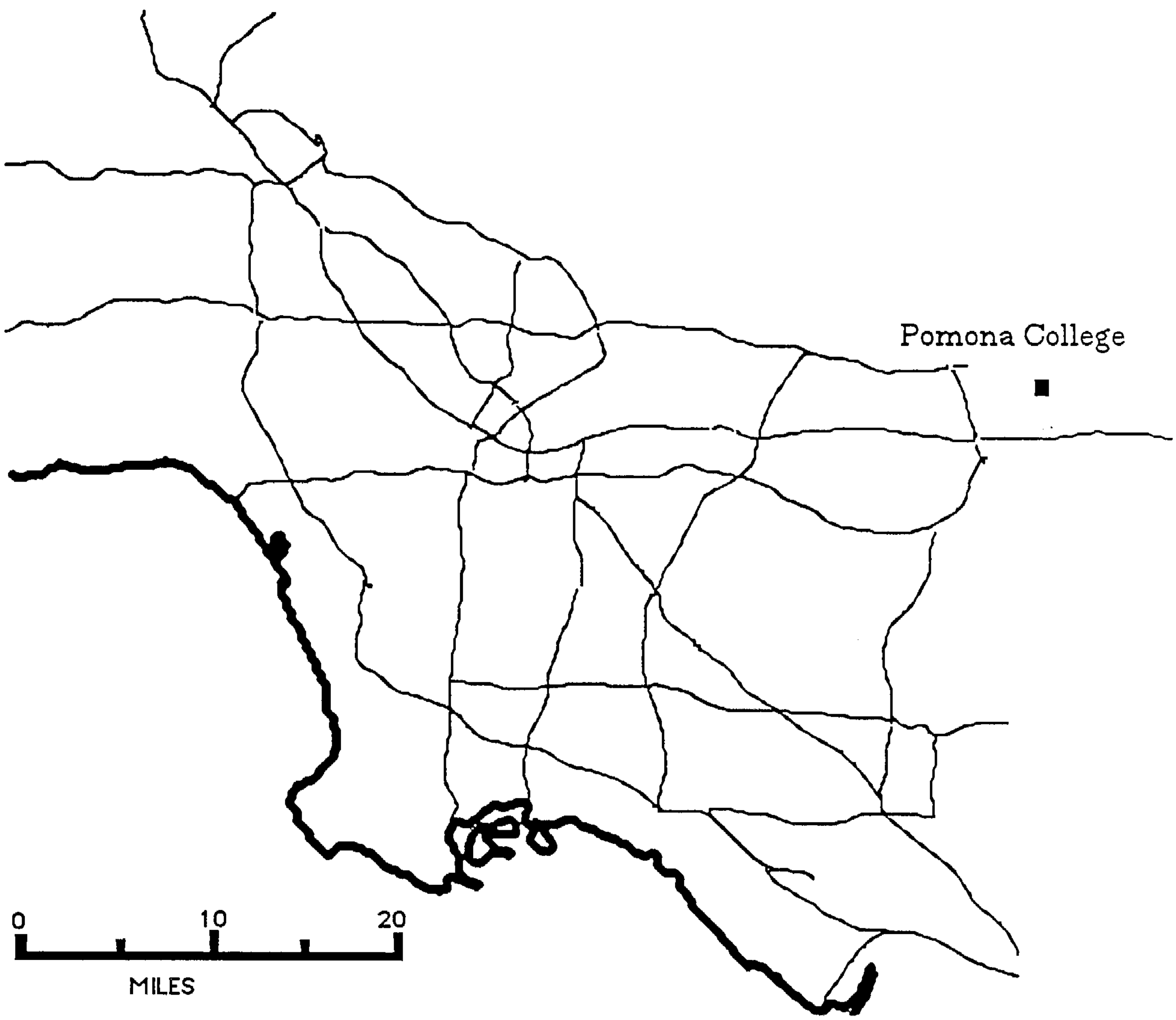

Figure 1. Map of the Los Angeles Basin showing the Pomona College sampling site in Claremont, CA.

two configurations is listed in Table 2, wherein locations are referenced with respect to the siting labels shown in Figure 2.

The sampling schedule was designed to accommodate the needs of both short-term and long-term samplers. It was felt each sampler should be tested in the mode for which it was designed. Thus investigators were given the choice of one of three schedules, consisting of five, two or one sampling period per day. Each investigator was asked to follow the same sampling schedule throughout the study. The five-per-day schedule consisted of four consecutive 4-hour periods starting at 0800 PDT, followed by one 6-hour period from midnight to 0600. These periods were selected to coincide with peak concentrations of various species during each day. The two-per-day schedule had one 12-hour sampling period from 0800 to 2000 and one 10-hour period from 2000 to 0600 the following morning. The one-per-day schedule ran from 0800 to 0600 the following morning. Samples were not collected between 0600 and 0800. This time was used for instrument calibration checks and the collection of field blanks. Some groups ran with identical samplers on different schedules to assess the effect of sampling duration.

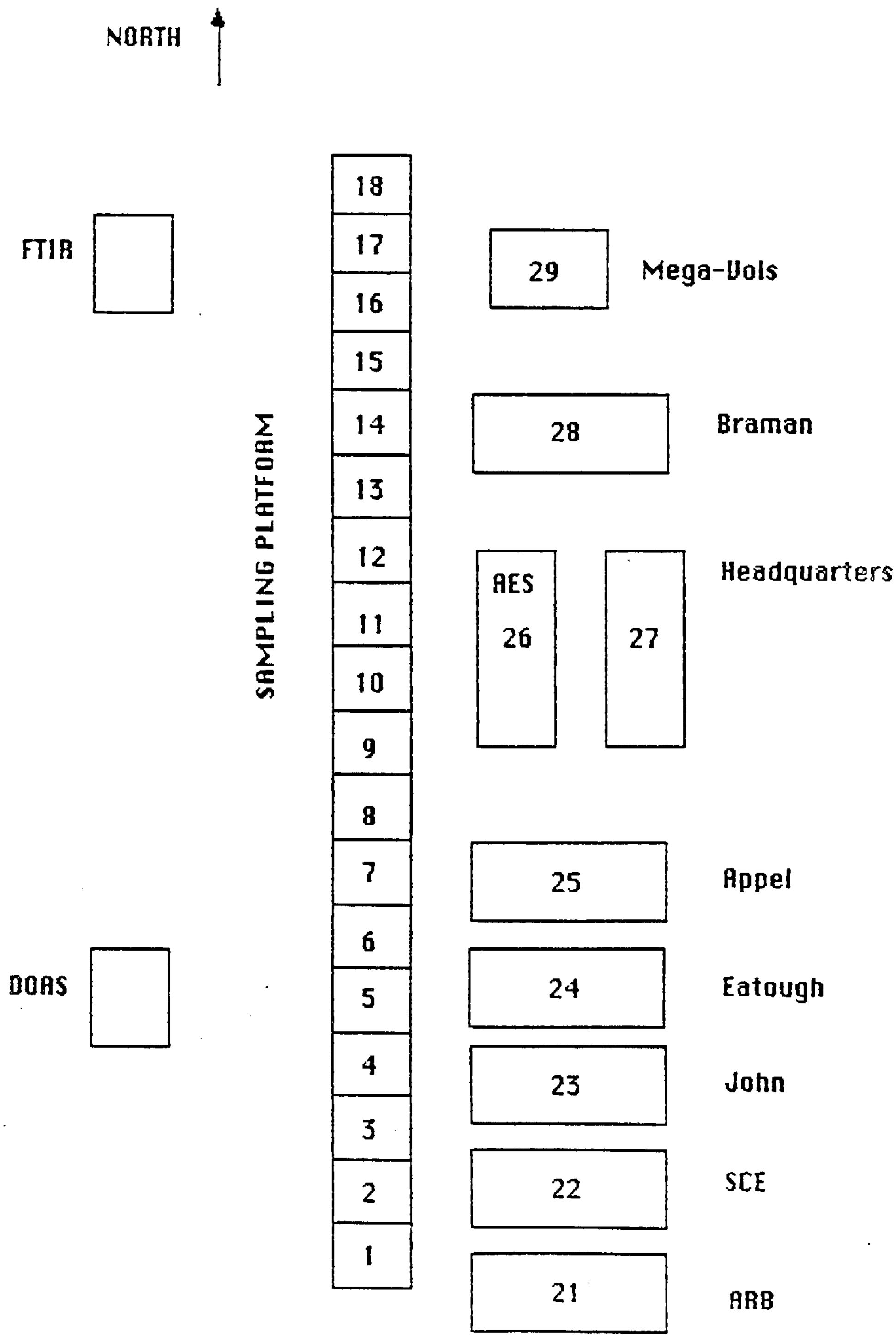

Figure 2. Diagram of the sampling site, with the siting labels used to identify sampler locations.

TABLE 2. SAMPLER SITING

	ID	SAMPLER	LOCATION 9/11-9/12	LOCATION 9/13-9/18
Filter Packs				
Anlauf/Wiebe	CF1	-T/N/Citric Acid (5/day)	1	1
	CF2	-T/N/Citric Acid (16/day)	1	12
Cass/Solomon	GF1	-T/N, -T/Ox/Ox	2	12
	GF2	-T/N, -T/Ox/Ox (1/day)	2	12
	GF3	-T/N, -T/Ox/Ox	2	1
	GF4	-T/N, -T/Ox/Ox: (network)	2	1
Grosjean	MF1	-N/N & -T/N	3	14
Martin/Mitchell	JF1	-T/N/K2CO3	4	4
	JF2	-T/N/K2CO3	4	4
Womack	NF1	El-T/N/S (2/day)	3	14
	NF2	El-T/N/S (6 days/sample)	3	14
	NF3	El-T/N/S (6 days/sample)	3	14
	NF4	-T/N/S (6 days/sample)	3	14
Denuder Difference Methods				
Appel	AD1	Cy-D(MgO)/N, -Cy-N	6	6
Cass/Solomon	GD1	Cy-{-D(MgO)/N, -N, -T}	10	10
	GD2	Cy-{-D(MgO)/N, -N, -T} (1/day)	10	11
	GD3	Cy-{-D(MgO)/N, -N, -T}	10	11
Horrocks	FD1	Cy-D(MgO)/T/N, -Cy-T/N	9	2
John	BD1	Dichot{T/N,T/N}	7	7
	BD2	Cy-T/N	7	7
	BD3	D(Anodized Al)/Dichot{T/N,T/N}	7	7
Pierson	KD1	Cy-{-D(MgO)/N, -N} -T	13	13
	KD2	Cy-{-D(MgO)/N, -N} -T (1/day)	13	13
Annular Denuders				
Sickles/	IA1	Cy-D(Na2CO3,Na2CO3,Citric Acid)/	18	9
Allegrini	IA2	/T/N/D(Citric Acid or KI&NaAsO2)	18	9
Eatough	EA1	Cy-D(Na2CO3,Na2CO3)/Q/N (4/dy)	16	16
	EA2	Cy-D(Na2CO3,Na2CO3)/Q/N (2/dy)	16	16
Peake	QA1	Cy-D(Na2CO3,Na2CO3,Citric Acid)/T/N	17	16
Transition Flow Reactors				
Ellestad	HE1	Cy-{-D(N,Nafion)/T/N/Ox/TEA/TEA	15	15
	HE2	-D(N,Nafion)/T/N/Ox/TEA/TEA}	15	15
Spicer	DE1	Cy-{-D(N,Nafion)/T/N/Ox/TEA/TEA	14	3
	DE2	-D(N,Nafion)/T/N/Ox/TEA/TEA}	14	3
Continuous Methods				
Stedman	SC1	Luminol HNO3	5	5
Winer/Tuazon	RC1	FTIR (HNO3, NH3)	30	30
Anlauf/Wiebe	CC1	T/TDLAS (HNO3, NO2)	26	26
Mackay	PC1	T/TDLAS (HNO3, H2CO)	26	26
Appel	AC1	Tungstic Acid Tubes	6	6
Braman	TC1	Tungstic Acid Tubes	28	28
Other Gaseous Species				
Appel	AO1	-D(Ox)	25	25
Appel	AF1	-T/Ox/Ox	6	6
Winer/Biermann	RC2	DOAS (HONO, NO3')	30	30
Spicer/Holdren	DC1	PAN - GC	21	21
Grosjean	MF2	-KOH/KOH & -DNPH/DNPH	14	14
Grosjean	MF3	-T/KOH & -T/DNPH	14	14
Mackay	PC2	Unisearch Luminox	26	26
Anlauf/Wiebe	CC2	T/N/NOx Box	26	26
Appel	–	Dasibi 1003	25	25
Ellis	–	Gases {O3, SO2, NOx,CO}	22	22
Eatough	–	GC-FPD (Dimethylsulfate)	24	24
Other Aerosol Measurements				
John	BO1	Berner Low Pressure Impactor	8	8
Ellis	OO1	Dichotomous Sampler	22	22
Lawson	FO1	HiVol/G	27	27
	FO2	HiVol/Q	27	27
	FO3	PM10-HiVol/Q	27	27
Meteorological Measurements				
Appel	–	Met {T, RH}	25	25
Ellis	–	Met {T, RH, WS, WD, P, Rad}	22	22
Special Experiments				
Eatough	EO3	Im-D(Ox,Ox,Ox,Ox,)/Q/N/Ox	16	16
	EO2	Im-{-D(TA,TA,TA,TA)/Q/N/Ox	16	16
	EO6	-Q/N}	16	16
	EO1	Im-{D(N,N,N,N,N,N,)/Q/N	16	16
	EO5	-D(N)/Q/N}	16	16
	EO7	-D(6xNa2CO3)/Q/N	16	16
Ellestad	HO1	Cy-D(N)/T/D(N)/N (4 days/dy-nt pr)	15	15
Filter Tests	TO	-T/N(old Membrana)	4	N/A
	TL	-T/N(new, low dP, #871)	4	N/A
	TH	-T/N(new, high dP, #4195)	4	N/A
Deposition Measurements				
John	BO2	Plants & Artificial Surfaces	8	8
Pierson	KO1	Dew Collectors	–	–
Pierson	KO2	Surrogate surfaces	–	–
Atmospheric Mutagens				
Winer/Atkinson	RX1	MegaVol/T impr.glass & G	29	29
	RX2	PM10-HiVol/T impr.glass	29	29
		PM10-HiVol/G	29	29
	RX3	Tenax columns	30	30

III. PRESENTATION OF RESULTS

Quality Control Checks: Quality Assurance Filters and Replicate Samplers

Quality Assurance Filters

Thirteen of the groups in the study relied on ion chromatographic analyses for integrated measurements of nitric acid and particulate nitrate. Since each laboratory analyzed its own filters, the consistency among laboratories was checked by asking each group to analyze a set of quality assurance filters upon which known amounts of sulfate and nitrate had been deposited. The quality assurance filters were prepared by Columbia Scientific Industries (Austin, TX). Each set consisted of nine Teflon® filters (1µm pore Zefluor, Gelman Sciences, Ann Arbor, MI) and nine nylon filters (1 µm Nylasorb, Gelman Sciences). Nitrate loadings on the Nylasorb filters, as specified by Columbia Scientific Industries, were 9.90 ± 0.01 µg, 69.0 ± 0.15 µg and 197.7±0.5 µg. The nitrate loadings on the Zefluor filters were 24.85±0.04 µg, 74.85±0.14 µg, and 249.3±0.6 µg. Laboratories were provided with three filters at each level of ion loading. Results from each laboratory are shown in Table 3 and Table 4. Data for each nitrate and sulfate loading are listed by the filter series number (100, 200 and 300).

For the nitrate analysis of the nylon filters, the average value from the analytical laboratories is generally in good agreement (≤4% deviation) with the deposited nitrate loadings, with the exception of the lightly loaded, series 100 Nylasorb filters. Except for groups B and C (who used large sampling volumes), the series 100 filters are representative of the amount of nitrate collected during many of the nighttime sampling periods. The accuracy of the analytical laboratories is presented in terms of the percent deviation from the deposited value.

For the filters loaded with 69 µg and 198 µg (series 200 and 300), 17 of 28 values agree within 3% of the deposited loading; all but one group report values within 11%. For filters loaded with 9.9 µg of nitrate (series 100), only 7 of the 14 groups report values within 8% of the deposited loading. The values from two laboratories are low at all three nitrate loadings. These two groups, N and I, used deionized water for the filter extractions, whereas all other laboratories used eluent or basic solution. For groups N and I, the sulfate levels for the lightly loaded Nylasorb are

Table 3. Nitrate results from quality assurance filters.

NYLON FILTERS (μg NO3-/filter)

GROUP	Series 100			Series 200			Series 300			Accuracy % Deviation from Spiked Value Ser 100	Ser 200	Ser 300	Measurment Precision Coefficient of Variation (%) Ser 100	Ser 200	Ser 300	Pooled
Womack	4.3	4.3	4.3	48.0	45.0	48.0	170	160	170	-57	-32	-16	0.0	3.7	3.5	2.8
Wiebe	9.8	10.0	10.1	67.0	67.7	67.3	198	199	198	01	-02	00	1.5	0.5	0.3	0.3
Spicer	13.0	13.2	13.6	80.5	80.5	78.3	216	217	199	34	16	06	2.3	1.6	4.8	3.4
Sickles	3.7	3.8	3.4	48.3	49.9	48.7	164	161	160	-63	-29	-18	5.7	1.7	1.3	1.1
Pierson	9.8	9.8	9.6	69.4	70.2	70.2	196	191	190	-02	01	-03	1.2	0.7	1.7	1.2
Peake	9.8	9.6	9.3	69.8	70.6	69.1	198	197	197	-03	01	00	2.6	1.1	0.3	0.4
Mitchell	0.0	2.3	0.0	67.8	65.8	69.0	207	190	187	-92	-02	-02	173.2	2.4	5.5	4.2
John	11.4	10.9	11.5	68.4	72.3	68.8	188	192	179	14	01	-06	2.9	3.1	3.6	2.6
Horrocks	10.3	10.1	9.6	71.9	76.7	68.4	178	176	175	01	05	-11	3.6	5.8	0.9	1.7
Grosjean	10.9	10.5	10.8	70.5	70.1	70.8	199	199	199	08	02	01	1.9	0.5	0.0	0.1
Ellestad	10.3	10.3	10.5	70.4	70.1	70.5	200	199	199	05	02	01	1.1	0.3	0.3	0.2
Cass/Solo	9.0	8.8	9.4	70.6	70.0	70.0	206	208	206	-08	02	04	3.4	0.5	0.6	0.4
Appel	6.5	6.9	7.8	67.0	68.0	67.0	193	195	195	-29	-02	-02	9.4	0.9	0.6	0.5
Eatough	11.9	12.1	12.0	68.7	68.7	69.2	185	184	184	21	00	-07	0.8	0.4	0.3	0.3
Average	8.7			67.1			191			-12	-03	-04				
Spike Val.	9.9	9.9	9.9	69.0	69.0	69.0	198	198	198							

TEFLON FILTERS (μg NO3-/filter)

GROUP	Series 400			Series 500			Series 600			Accuracy % Deviation from Spiked Value Ser 400	Ser 500	Ser 600	Measurment Precision Coefficient of Variation (%) Ser 400	Ser 500	Ser 600	Pooled
Womack	26.0	24.0	26.0	69.0	67.0	69.0	220	240	260	02	-09	-04	4.6	1.7	8.3	6.0
Wiebe	24.6	24.3	24.5	72.6	72.6	73.7	256	247	253	-02	-03	01	0.6	0.9	1.8	1.3
Spicer	27.3	27.3	26.9	79.3	80.0	80.0	269	265	275	09	06	08	0.9	0.5	1.9	1.3
Sickles	24.4	24.2	24.8	73.9	73.5	73.6	247	250	248	-02	-02	00	1.2	0.3	0.6	0.5
Pierson	25.2	23.8	22.8	77.4	76.4	72.1	252	257	247	-04	01	01	5.0	3.7	2.0	1.7
Peake	25.8	25.4	25.3	74.7	76.5	75.9	254	254	279	02	01	05	1.0	1.2	5.5	4.0
Mitchell	26.8	23.8	14.8	62.3	58.0	71.3	253	250	251	-12	-15	01	28.6	10.6	0.6	2.8
John	25.8	25.8	25.7	71.8	78.0	78.7	221	238	246	03	02	-06	0.2	5.0	5.4	4.0
Horrocks	29.6	30.5	30.0	99.3	107.5	113.8	226	239	245	21	43	-05	1.5	6.8	4.1	3.2
Grosjean	26.6	26.2	26.3	77.6	77.0	77.5	263	257	260	06	03	04	0.8	0.4	1.2	0.8
Ellestad	25.0	25.1	25.0	75.6	76.1	75.5	250	251	250	01	01	01	0.2	0.4	0.2	0.2
Cass/Solo	23.9	20.2	19.8	76.5	77.3	77.8	264	261	240	-14	03	02	10.6	0.8	5.1	3.8
Appel	22.0	22.0	22.0	79.0	79.0	79.0	233	236	231	-12	05	-06	0.0	0.0	1.1	0.8
Eatough	26.7	69.2	25.6	26.8	74.6	75.3	221	228	229	63	-21	-09	61.4	47.1	1.9	11.5
Average	26.0			75.8			248			04	01	00				
Spike Val.	24.9	24.9	24.9	74.9	74.9	74.9	249	249	249				25	75	249	

Table 4. **Sulfate results from quality assurance filters.**

NYLON FILTERS (µgSO4--/filter)

GROUP	Series 100			Series 200			Series 300			Accuracy % Deviation from Spiked Value Ser 100	Ser 200	Ser 300	Measurment Precision Coefficient of Variation (%) Ser 100	Ser 200	Ser 300	Pooled
Womack	9.0	8.0	9.0	88.0	87.0	90.0	330	230	240	-56	-11	08	6.7	1.7	20.7	15.2
Wiebe	20.1	19.9	20.1	103.0	100.0	100.0	262	263	268	01	02	07	0.6	1.7	1.2	0.9
Spicer	20.9	23.1	23.6	110.3	110.3	109.0	263	263	250	14	11	05	6.4	0.7	2.9	2.0
Sickles	6.9	9.6	8.0	90.5	89.3	90.6	236	236	235	-59	-09	-05	16.3	0.8	0.2	0.5
Pierson	19.7	19.4	19.2	100.0	101.1	101.6	250	247	246	-02	02	00	1.3	0.8	0.8	0.6
Peake	18.1	17.7	18.3	93.9	94.9	93.1	232	234	233	-09	-05	-06	1.7	1.0	0.4	0.4
Mitchell	19.5	18.0	17.0	103.0	97.0	104.0	235	230	221	-08	03	-07	6.9	3.7	3.1	2.3
John	18.2	18.2	19.5	94.3	99.6	94.2	237	237	227	-06	-03	-05	4.0	3.2	2.5	1.9
Horrocks	19.0	20.1	19.2	98.9	114.0	99.1	228	214	205	-02	05	-13	3.0	8.3	5.3	4.2
Ellestad	20.2	20.2	20.1	94.5	95.8	96.1	245	244	245	02	-03	-01	0.3	0.9	0.2	0.3
Eatough	22.4	22.8	22.6	96.8	95.6	96.8	205	203	203	14	-02	-17	1.1	0.7	0.6	0.4
Cass	19.7	20.1	20.0	103.0	101.0	101.0	262	255	254	01	03	04	1.0	1.1	1.7	1.2
Average	18.0			98.3			241			-09	00	-03				
Spike Val.	19.8	19.8	19.8	98.7	98.7	98.7	247	247	247							

TEFLON FILTERS (µgSO4--/filter)

GROUP	Series 400			Series 500			Series 600			Accuracy % Deviation from Spiked Value Ser 400	Ser 500	Ser 600	Measurment Precision Coefficient of Variation (%) Ser 400	Ser 500	Ser 600	Pooled
Womack	16.0	15.0	16.0	46.0	47.0	48.0	350	250	230	05	-06	24	3.7	2.1	23.2	18.9
Wiebe	14.9	15.0	15.0	48.2	48.5	49.4	233	228	229	00	-02	03	0.4	1.3	1.2	0.9
Spicer	18.1	17.9	18.4	52.8	54.5	54.5	241	236	241	22	08	07	1.4	1.8	1.2	1.0
Sickles	14.8	14.8	14.9	49.0	49.5	49.6	215	223	222	-01	-01	-02	0.4	0.7	2.1	1.7
Pierson	15.0	14.5	14.2	51.1	49.9	47.7	227	230	224	-02	-01	01	2.8	3.5	1.4	1.2
Peake	15.1	15.2	14.9	51.2	49.5	49.1	225	220	218	01	00	-01	1.0	2.2	1.6	1.3
Mitchell	15.5	14.8	12.5	47.0	44.3	53.0	213	213	215	-04	-04	-05	11.0	9.3	0.5	1.8
John	14.0	13.8	13.8	44.9	49.5	49.6	196	211	220	-07	-04	-07	0.8	5.6	5.7	4.5
Horrocks	16.0	18.0	18.5	66.2	72.0	72.1	210	218	222	17	40	-03	7.6	4.8	2.8	2.3
Ellestad	14.9	14.9	14.9	48.8	49.2	49.7	219	221	220	00	-01	-02	0.0	0.9	0.5	0.4
Eatough	16.3	15.4	16.3	51.1	52.1	52.7	182	187	187	07	04	-17	3.5	1.6	1.5	1.1
Cass	13.8	13.0	14.6	51.9	50.6	51.1	245	231	211	-07	03	02	5.8	1.3	7.5	5.8
Average	15.3			51.4			224			03	03	00				
Spike Val.	14.9	14.9	14.9	49.9	49.9	49.9	224	224	224							

also low; however, their results for both nitrate and sulfate on the Zefluor (Teflon) filters are within 2% of the deposited value in one case, and within 15% in the other.

The precision of the individual laboratories is given by the coefficient of variation for analyses of 3 filters at each level of deposited nitrate. Generally the precision of the laboratories is good. For the series 200 and 300 filters, coefficients of variation are less than 6% for all fourteen laboratories, and nine of the laboratories have coefficients of variation less than 1%. For the series 100 filters (with 9.9μg of deposited nitrate), twelve laboratories report standard deviations of less than 0.4 μg NO_3^-/filter. Analytical precision, calculated using two times the pooled standard deviation divided by the appropriate sample volumes for each group, is better than 25 neq/m3 for all groups except D, J and N .

Some of the nylon filters used by participants were older filters manufactured by Membrana (Pleasanton, CA). Others were newer filters purchased from Gelman, following the sale of the Membrana company to Gelman. The newer Gelman filters were somewhat different in appearance, and often had a larger pressure drop for the same face velocity. The old Membrana filters and two lots of the new Gelman filters were compared in side-by-side sampling with filter packs (sampler nos. TO, TL, and TH in Table 1). No differences were observed in the collection efficiency for HNO_3, although the new filters did retain some SO_2.

Replicate Samplers

Before assessing the differences among samplers, we must first ask how precise the measurement is for a specific sampler type. There were five pairs of replicate samplers in the study. Two of these like sampler pairs, viz. the CIT filter packs #GF1 and #GF3 and the Canadian filter packs #CF1 and #CF2, were located side by side for the first two days of the study, and then were sited at different locations on the platform for the remainder of the study. The other three like sampler pairs were collocated throughout the study. The collocated measurements are indicative of the overall precision, including both sampling and analysis, which can be expected from the method, whereas the separated measurements include the increased variability due to sampler siting.

For collocated sampling, the standard deviations within sampler pairs are, respectively, 15 neq/m^3, 49 neq/m^3, 15 neq/m^3, and 25 neq/m^3 for filter packs #GF1 and #GF3, denuder difference samplers #GD1 and #GD3, annular denuders #IA1 and #IA2, and TDLAS systems #CC1 and #PC1. The corresponding coefficients of variation are 12%, 27%, 35% and 20%.

Data from the separated CIT filter packs #GF1 and #GF3 are shown by the solid symbols in Figure 3. A linear least squares regression gives a slope of 1.03 and a correlation coefficient of 0.99, indicating no systematic bias due to sampler location on the platform. The coefficient of variation between the two samplers is 12%, which is the same value observed when the filter packs were collocated, and approximates the precision expected on the basis of the CIT quality assurance filter data and sample volumes. Similarly, the coefficient of variation between the separated Canadian filter packs was 15%. Therefore, the siting of the samplers on the platform did not introduce a systematic bias, nor did it contribute significantly to the scatter in the data.

Ambient Nitric Acid Measurements

Time series plots of the nitric acid concentrations measured during the study are shown in Figures 4 and 5. These are grouped according to measurement method, with filter pack data and denuder difference method results in Figure 4 and the annular denuders, the transition flow reactors, diode lasers and the tungstic acid hollow tube methods in Figure 5. (FTIR data are available for a only small part of the study, and are presented in a later section.) The data all show the same trends, with clear diurnal variations in the nitric acid levels and peak daytime nitric acid concentrations on September 14, 1985.

Several results are apparent from inspection of the data. For the most part, the variation among sampling periods is greater than the differences among measurement methods. The highest reported nitric acid concentrations are from the filter packs. For example, on the high nitric acid day of September 14, values reported by most filter packs are higher than those observed by the diode laser systems. Results from the denuder difference methods on this day are intermediate.

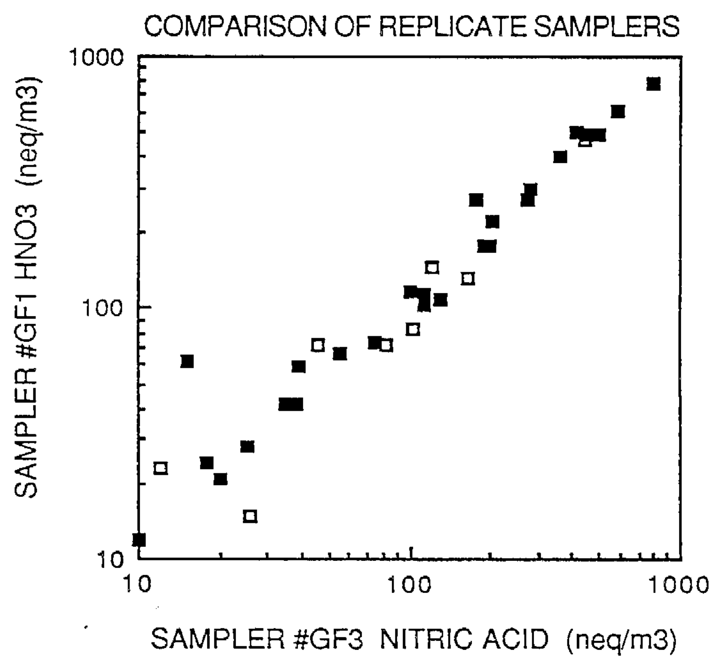

Figure 3. Comparison of simultaneous 4- and 6- hour nitric acid data obtained from two filter packs of the same design, operated by the California Institute of Technology. Data are shown for collocated (□) and separated (■) siting of the samplers along the instrument platform.

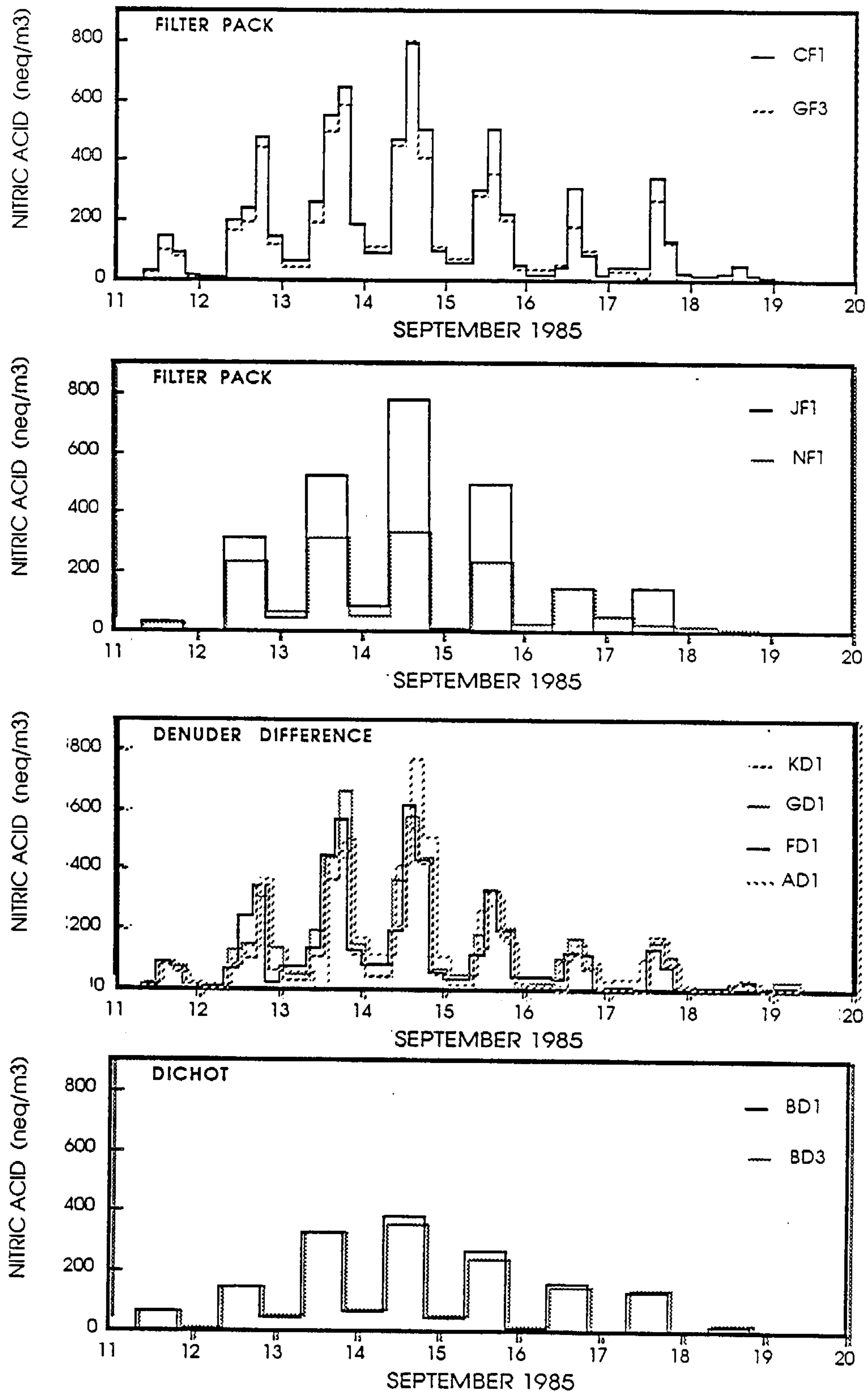

Figure 4. Time series plots of nitric acid data from filter packs and denuder difference samplers for 10- and 12-hour sampling, and for 4- and 6-hour sampling. Sampler configurations and identification numbers are given in Table 1.

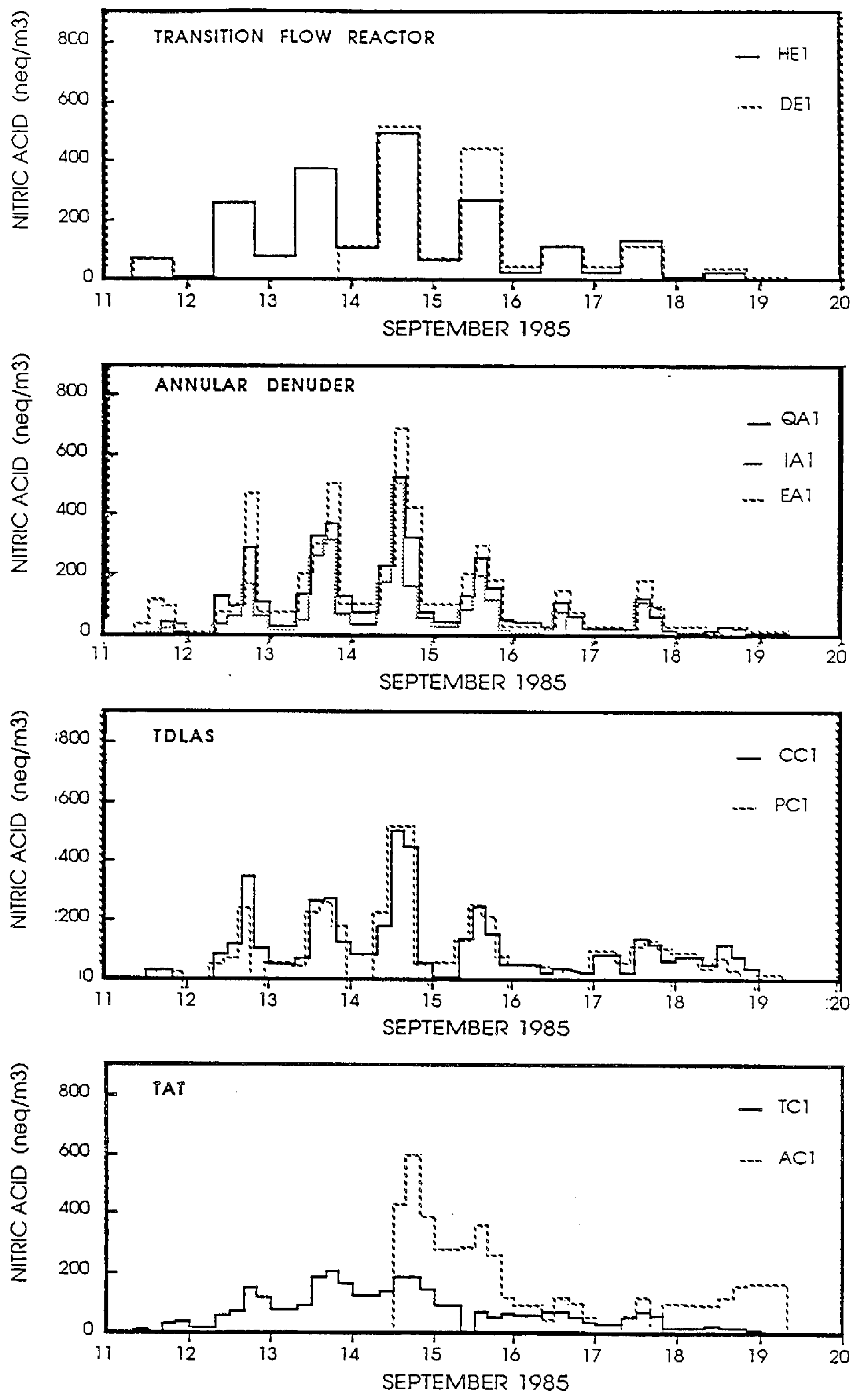

Figure 5. Time series plots for nitric acid data from transition flow reactors, annular denuders, tunable diode laser absorption spectrometers, and tungstic acid technique systems. Sampler identification numbers are given in Table 1.

Comparing samplers within each sampler type, we find that the 4- and 6-hour filter pack samplers agree reasonably well. The same is true of the denuder difference samplers, the two transition flow reactors and the two laser diode spectrometers. There is somewhat more scatter among the three annular denuders. Large discrepancies are observed between the two tungstic acid technique systems. These were not identical systems. The two TAT samplers were constructed and operated by different groups. While using the same principle for nitric acid detection, the systems differed in their inlet design and in the operational procedure.

The standard deviation in the reported nitric acid concentration for each sampling period among all of the instruments shown in Figures 4 and 5 is plotted in Figure 6. For both the 10- and 12-hour sampling and the 4- and 6-hour sampling the standard deviation increases linearly with the nitric acid concentration. In both cases the data can be described well by a coefficient of variation of 40%. The corresponding range in reported nitric acid concentrations in a given sampling period is as much as a factor of four. For example, on the afternoon of September 14 (period 43) reported nitric acid concentrations varied from 190 neq/ m^3 to 800 neq/ m^3, with all but two samplers giving values between 330 and 800 neq/m^3. The differences among samplers are larger than can be explained by the analytical accuracy as indicated by the quality assurance filter analysis, or by the overall sampling precision as indicated by the replicate samplers.

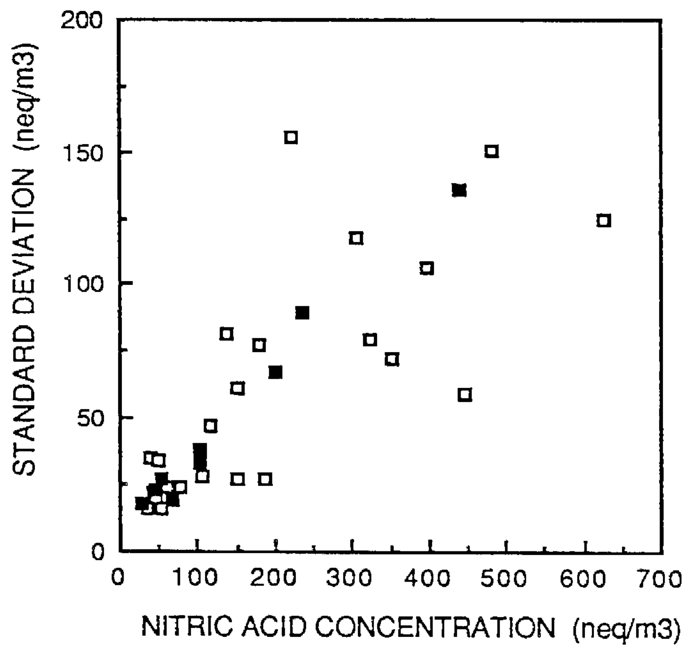

Figure 6. The standard deviation in the reported nitric acid for all samplers is shown as a function of the average nitric acid concentration for each of the sampling periods. Four- and six-hour sampling is shown by the open symbols (□), ten- and twelve-hour sampling is shown by the solid symbols (■).

IV. ANALYSIS OF NITRIC ACID DATA

Approach

Questions addressed in this data analysis are : (1) the systematic differences among individual samplers and among sampling methods, (2) the measurement precision for each sampler, and (3) diurnal variations in sampler performance. Two sets of analyses are presented. First the samplers are compared individually, and second, the different measurement methods (e.g. FP vs DDM) are compared. Although the differences between types of methods may be of greatest interest, it is necesary to compare the samplers individually to avoid prejudging the data. The individual sampler comparisons also provided a basis for selecting representative samplers for the intermethod comparisons. Thus the comparison between measurement methods presented here is based on a subset of the data which excludes samplers with imprecise data.

Comparisons Among Individual Samplers: 10- and 12-hour Data

To compare the different samplers we have selected a subset of the data for which we have a complete data set. Thus four of the samplers are excluded from this analysis: the Luminol semicontinuous nitric acid monitor, one TFR (#DE1), one TAT system, (#AC1), and the FTIR. Replicate samplers of the same design operated by the same group were not included. The data from those samplers collecting 4 and 6 hour samples have been composited to provide 10- and 12- hour averages for comparison with the half day collection periods.

A total of 18 samplers are compared over nine half-day sampling periods (nos. 16, 26, 27, 37, 46, 47, 56, 57 and 76, as given in Table A2) for which a complete data set was available, and for which median concentrations were above 20 neq/m^3. Samplers with 10- and 12-hour sampling times include two filter packs (#JF1 and #NF1), two dichotomous denuder difference samplers (#BD1 and #BD3), and one transition flow reactor (#HE1). Samplers operating on the 4- and 6-hour sampling times, whose data are composited for this comparison, include two filter packs, #CF1 and #GF3, four denuder difference samplers, #AD1, #FD1, #GD1 and #KD1, and two annular denuders, #IA1 and #QA1. Continuous data are obtained from the two tunable diode laser absorption spectrometers, #CC1 and #PC1, and from the tungstic acid hollow tube system #TC1. One filter pack, #EF1, and one annular denuder, #EA1, operated on the 4-hour schedule during the day, and the 10-hour schedule at night. The periods considered include five daytime samples and four nighttime samples. The mean nitric acid concentrations range from 30 to 440 neq/m^3, mean temperatures vary from 17^{o}C to 28^{o}C and mean relative humidities are 30% to 90%.

In this analysis the samplers are considered individually without grouping them by the measurement method. Systematic differences among the samplers are evaluated using the Friedman analysis of variance by rank, which is a distribution-free, non-parametric statistical test (Hollander and Wolfe, 1973). Nitric acid concentrations from each sampler are ranked from highest to lowest for each period, and then the samplers are compared on the basis of their average ranking. One advantage of this particular statistical test is that it does not assume that the data are normally distributed. Each sampling period is given equal weight in the analysis.

The Friedman analysis of variance shows there are significant differences among the samplers at a >99.9% confidence level. The average ranking of the samplers, based on the reported nitric acid concentrations, is shown in Figure 7. The highest concentrations are from the filter packs #CF1 and #GF3. The lowest concentrations are from one of the annular denuders, #IA1. Pairwise comparisons, based on the Friedman rank sums, show that, at the 95% confidence level, the nitric acid concentrations reported by #IA1 are lower than those reported by the #CF1 and #GF3 filter packs and the #HE1 transition flow reactor:

#IA1 < #CF1
#IA1 < #GF1
#IA1 < #HE1

No statistically significant difference is seen among the other samplers with the Friedman analysis.

To assess measurement precision, we work with the log transformation of the data. The data are much better approximated by a lognormal distribution than by a normal distribution. However, the lognormal distribution is a poor assumption for the low concentration data. When the nitric acid concentrations are less than 25 neq/m^3 (≈0.6 ppb), analytical errors are significant, and therefore these periods are not included in the analysis.

The precision of each of the measurement techniques is assessed by calculating the mean squared residual for each sampler j:

$$R_j = (1/n)\,\Sigma_i(\mathrm{Log}(S_{ij}/M_i))^2 \qquad (1)$$

where S_{ij} is the nitric acid concentration reported by sampler j for period i, and M_i is the geometric mean concentration reported by all 18 samplers for period i, and n is the number of periods. The mean squared residual is the average of the squares of the difference between the logarithm of the sampler nitric acid concentration and the logarithm of the geometric mean concentration among all samplers for the sampling period. It reflects both the random and systematic differences between the sampler and the mean.

Results for the eighteen samplers evaluated for the half-day sampling periods are presented in Figure 7. Because of the logarithmic transformation of the data, the mean squared residual does not give error bounds in terms of neq/m^3 nitric acid, nor does it correspond to a coefficient of variation. However, the relative comparison between samplers is meaningful. We expect the samplers whose nitric acid concentrations are systematically high or systematically low (at the extreme ends of the ordinate in Figure 7) to have somewhat higher residuals than those samplers in the middle because of the greater systematic difference from the mean.

Two of the samplers, the filter pack #JF1 and the TAT sampler #TC1, have very large mean squared residuals. When the daytime and nighttime data are examined separately, we find that the nitric acid concentrations reported by the TAT sampler #TC1 are consistently high at night and low during the day. The opposite is true for the filter pack #JF1. The diurnal trends in the data from these two samplers can also be seen by close inspection of the time series plots of Figures 2 and 3.

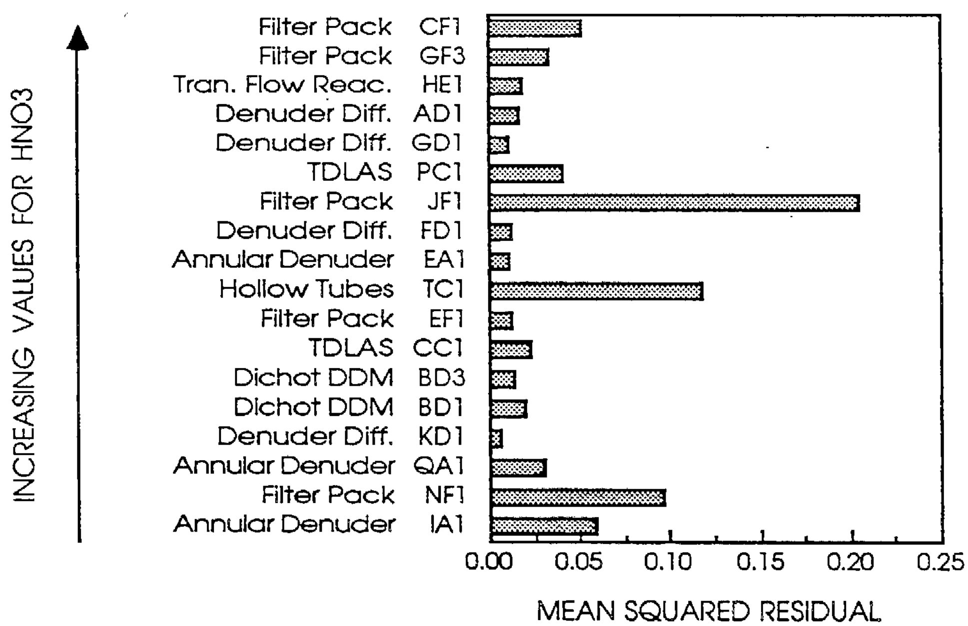

Figure 7. The measurement precision, as given by the mean squared residual (eqn. 1) is shown for eighteen of the samplers. Samplers are listed according to average rank, with those giving the highest nitric acid levels at the top, lowest at the bottom.

Finally, we wish to quantify the differences among those samplers with relatively small values for the mean squared residual. One possible parameter is the ratio in nitric acid concentrations between samplers. We first examine whether the ratios are a reasonable characterization of the differences between samplers. As an example, in Figure 8 the ratios for samplers #BD1 and #QA1 to the sampler #HE1 are plotted as a function of the geometric mean nitric acid concentration for each period. The ratios are plotted on a logarithmic scale so that reciprocal ratios (e.g. 2 and 1/2) are equally spaced around the value 1. We find these ratios to be randomly distributed with respect to the nitric acid concentrations, indicating that the ratios are a consistent way to represent the difference between samplers. Although not shown, the ratios between other samplers also appear to be random with respect to nitric acid levels.

As an unbiased estimator of the average ratio between samplers, we have calculated the ratio of mean values, or simply the ratio of the sum of nitric acid concentrations for all the periods considered. These data are given in Table 5. The samplers included in this analysis are the transition flow reactor #HE1, and the denuded and undenuded dichotomous samplers #BD1 and #BD3, all of which operated on the 10- and 12-hour sampling schedule. Samplers #EF1 and #EA1 collected three samples during the daytime hours, but only one sample at night, and thus can be compared only on the half-day sampling schedule. The other two annular denuders are included for comparison with the annular denuder #EA1. To provide a comparison with the samplers operating on the 4- and 6-hour schedule (Table 6), we have also included the ratios of means of the individual samplers to the mean from the four denuder difference method samplers (DDM). As discussed below, these values parallel the overall sampler mean and the FTIR data.

The sampler pairs marked by asterisks give significantly different results, based on Wilcoxon signed rank tests. This is a non-parametric test used for paired data (Hollander and Wolfe, 1973). It is used to indicated systematic differences between methods. By this test, only the annular denuder #IA1 and the transition flow reactor #HE1 were found to differ significantly. Both the denuded and undenuded dichotomous samplers, #BD3 and #BD1, give statistically equal results. The nitric acid values reported by the transition flow reactor are higher than those from the denuder difference samplers and annular denuders, but are somewhat

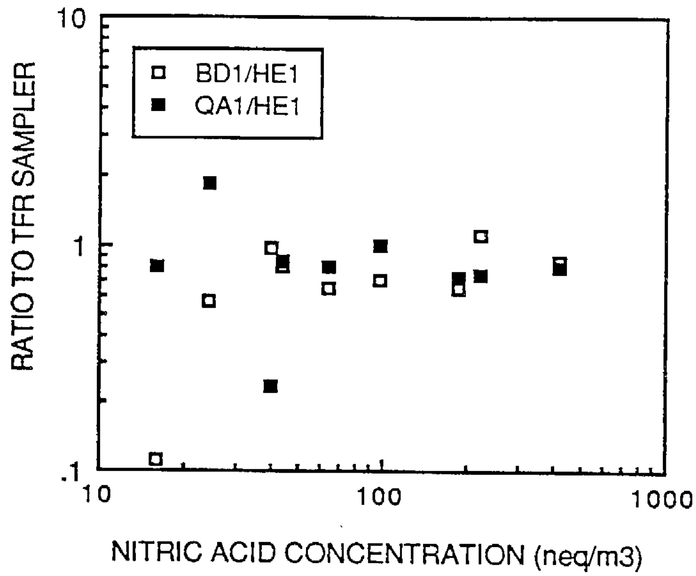

Figure 8. Concentration ratios between the dichotomous denuder difference sampler #BD1 and the transition flow reactor #HE1, and between the annular denuder #QA1 and the transition flow reactor #HE1, are plotted against nitric acid concentration. All data are for 10- and 12-hour sampling periods.

Table 5. Ratio of means between samplers for nitric acid for 10- and 12-hour sampling periods.

		$\Sigma y/\Sigma x$							
	x:	HE1	BD1	BD3	EF1	EA1	QA1	IA1	DDM†
y:	HE1	1.00	1.10	1.13	1.24	1.01	1.31	1.74 *	1.04
	BD1	0.91	1.00	1.03	1.13	0.92	1.19	1.58	0.94
	BD3	0.88	0.97	1.00	1.10	0.89	1.15	1.53	0.92
	EF1	0.81	0.89	0.91	1.00	0.82	1.05	1.40	0.84
	EA1	0.99	1.09	1.12	1.22	1.00	1.29	1.71	1.02
	QA1	0.76	0.84	0.87	0.95	0.78	1.00	1.33	0.80
	IA1	0.57 *	0.63	0.65	0.71	0.58	0.75	1.00	0.60
	DDM†	0.96	1.06	1.09	1.20	0.98	1.26	1.67	1.00

*Samplers differ at >95% confidence level by Wilcoxon signed ranks (see text).
†Average of denuder difference method samplers #AD1, #FD1, #GD1 and #KD1

lower than the #GF3 filter pack.

Comparisons Among Individual Samplers: 4- and 6-hour Data.

We compare here nine samplers which collected four 4-hour samples and one 6-hour sample per day. The set of samplers includes two filter packs, #CF1 and #GF3, four denuder difference methods, #AD1, #FD1, #GD1 and #KD1, one annular denuder #QA1, and the two diode laser spectrometers, #CC1 and #PC1. The TAT system #TC1 and the annular denuder #IA1 are not included. The samplers are compared over 15 daytime and 6 nighttime sampling periods (nos. 14, 22-24, 31, 33-35, 42-45, 51-55, 61, 63, 73 and 74, as listed in Table A1). For these periods geometric mean nitric acid concentrations ranged from 30 to 616 neq/m^3. Periods with geometric mean concentrations below 25 neq/m^3 (0.6 ppb) are not included in the analysis.

The mean squared residuals for the individual samplers, calculated from eqn (1), are all below 0.05, and are in approximate agreement with the values given in Figure 7. As with the 10- and 12-hour data, the ratios of the nitric acid concentrations reported by different samplers are not dependent upon the nitric acid concentrations, as shown for samplers #CF1, #FD1 and #GF3 in Figure 9. The characteristic ratio between sampler pairs is estimated by the ratio of means, as given in Table 6. The comparison with the average of the denuder difference method samplers (DDM) is also shown, and provides a basis for comparison with the data of Table 5.

In Table 6 we have also indicated which sampler pairs differ significantly at the 95% confidence level, using the Wilcoxon signed rank test. Significant differences are reported when individual Wilcoxon tests indicate a difference at the >99.86% confidence level, which for the 36 tests run here corresponds to an overall confidence level of 95%. This test is used to indicate whether one sampler consistently gives higher (or lower) results than the other. Whether or not two samplers prove different depends on the scatter in the data, as well as the magnitude of the difference.

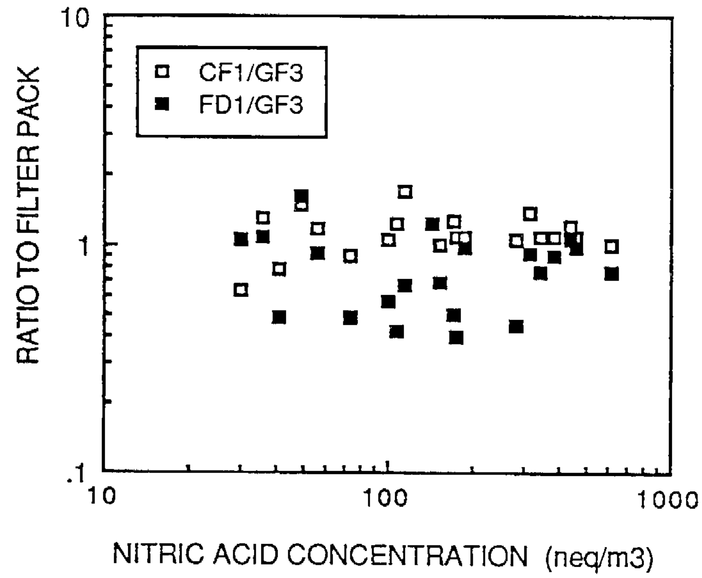

Figure 9. Concentration ratios between the Canadian filter pack #CF1 and the Caltech filter pack #GF3, and between the denuder difference sampler #FD1 and the filter pack #GF3 are plotted against nitric acid concentration. All data are for 4- and 6-hour sampling periods.

Table 6. Ratio of means between samplers for nitric acid for 4- and 6-hour sampling periods.

	$\Sigma y/\Sigma x$									
x:	CF1	GF3	AD1	FD1	GD1	KD1	QA1	CC1	PC1	DDM†
y: CF1	1.00	1.13*	1.31*	1.45*	1.33*	1.60*	1.79*	1.82*	1.78	1.41
GF3	0.88*	1.00	1.16*	1.28*	1.18	1.41*	1.59	1.60	1.57	1.25
AD1	0.76*	0.86*	1.00	1.11	1.02	1.22	1.37*	1.38	1.36	1.08
FD1	0.69*	0.78*	0.90	1.00	0.92	1.10	1.24	1.25	1.22	0.97
GD1	0.75*	0.85	0.98	1.09	1.00	1.20*	1.34*	1.36	1.33	1.06
KD1	0.63*	0.71*	0.82	0.91	0.83*	1.00	1.12	1.14	1.11	0.89
QA1	0.56*	0.63*	0.73*	0.81	0.74*	0.89	1.00	1.01	0.99	0.79
CC1	0.55*	0.62	0.72	0.80	0.73	0.88	0.99	1.00	0.98	0.78
PC1	0.56	0.64	0.74	0.82	0.75	0.90	1.01	1.02	1.00	0.80
DDM†	0.71	0.80	0.93	1.03	0.94	1.13	1.27	1.28	1.26	1.00

*Samplers differ at >95% confidence level by Wilcoxon signed ranks (see text).
†Average of denuder difference method samplers #AD1, #FD1, #GD1 and #KD1

On the average, the highest nitric acid concentrations are reported by the filter pack #CF1. Its values are significantly higher than those from the other filter pack #GF3. Both filter packs (#GF3 and #CF1) give significantly higher nitric acid levels than the denuder difference and annular denuder samplers. Among the denuder difference samplers, the ratios of means are between 0.8 and 1.2, but only for one pair, #GD1 and #KD1, did the difference prove to be statistically significant. The tunable diode laser absorption spectrometers reported lower values, on the average, than the denuder difference samplers and filter packs. The corresponding ratios of means to the TDLAS are between 1.1 and 1.8. However, the scatter of the data in the comparison of other samplers is such that the only the differences in sampler pair #CF1 and #CC1 were found to be statistically significant. The annular denuder #QA1 also gave lower results than the filter packs and denuder difference samplers, and gave the same average value as the tunable diode laser absorption spectrometers.

Comparisons Within Types of Measurements

The analysis of individual samplers shows that there can be differences among samplers using the same method, as well as among methods. For the filter packs #CF1, #GF3, #JF1,#EF1 and #NF1, the Friedman analysis of variance shows differences at the >99% confidence level. When comparing daytime and nighttime sampling periods, sampler #JF1 is high during the day and low at night. The Friedman analysis of variance shows no significant differences among the six denuder difference samplers, #AD1, #BD1, #BD3, #FD1, #GD1 and #KD1, compared over the 10- and 12-lhour sampling periods.

Using the Friedman analysis of variance, the nitric acid concentrations measured by the three annular denuders, #EA1, #IA1 and #QA1, differ at >99% confidence level. Pairwise comparisons using Friedman rank sums show (at the 95% confidence level) that #IA1 < #QA1 < # EA1. The differences are observed for both daytime and nighttime sampling.

Figure 4 shows that the two tunable diode laser absorption spectrometers and the two transition flow reactors agree fairly well within methods, while the two TAT systems give quite different results. Each pair of samplers is compared using the Wilcoxon signed rank test. We find no significant differences at the 95% confidence level between the two transition flow reactors and no significant differences between the two TDLAS systems. The two TAT systems differ at the 95% confidence level.

Comparison of 22-hour samples with the concurrent sum of 5-per-day samples reveals a potential limitation of the denuder difference method, namely, that the nylon filters become saturated at HNO_3 doses beyond approximately 30 µg per cm^2 of filter area. This saturation effect was first reported by Anlauf *et al.* (1986) in another setting. In the present instance, four denuder difference systems were compared, all employing 47 mm diameter nylon filters: (1) #KD1, collecting 5-per-day samples at flows of 10 and 18 L/min. through denuded and undenuded branches; (2) #GD1 and #GD3, 5-per-day samples but with flows 5 times less; (3) #KD2, 22-hour samples with flows of 10 and 18 L/min.; and (4)#GD2, 22-hour samples with flows five times less. Good agreement is consistently obtained between (1), (2) and (4). At low HNO_3 concentrations, agreement with (3) was good also. But on days 3 and 4, when total HNO_3 exposures of the filter on the undenuded side of (3) approached or exceeded 30 µg/cm^2, the results from (3) were low, reaching a 37% deficit at an exposure of approximately 35 µg/cm^2. These results indicate that HNO_3 exposures of greater than 30 µg/cm^2 must be avoided for proper use of the denuder difference method.

Comparisons by Method with the FTIR

In the preceeding sections no comparison is made with the FTIR data because complete data were obtained for only one of the 4-hour sampling periods. During other periods either the nitric acid concentrations were below detection limit, or the FTIR spectrometer was subject to noise interferences. Nevertheless, the FTIR data are of interest because it was the only instrument which did not use sampling lines or filters. Unlike the situation in the 1979 comparison study (Spicer et al. 1982), the optical path for the FTIR in this study was not enclosed within any cell walls. Accordingly, we assume that the FTIR measurements should be less subject to sampling artifacts, and thus comparisons with the other methods are of definite interest.

Rather than comparing individual samplers with the FTIR, we compare the averages of representative samplers for each of the measurement techniques. Values for filter packs (FP), denuder difference methods (DDM), annular denuders (ADM), transition flow reactors (TFR), tunable diode laser absorption spectrometers (TDLAS), and the dichotomous sampler denuder difference method (Dichot) are calculated as follows:

FP = 1/2 { #CF1 + 1/2(#GF1+#GF3) }
DDM = 1/4 { #AD1 +# FD1 + 1/2(#GD1+#GD3) + #KD1 }
ADM = 1/3 { #EA1 +# QA1 + #IA1 }
TFR = 1/2 { #HE1 + #DE1 }
TDLAS = 1/2 { #PC1 +# CC1 }
Dichot = 1/2 { #BD1 + #BD3 }

The filter packs are represented by samplers #CF1, #GF1 and #GF3 because each of these samplers used an open face Teflon® filter followed by a nylon filter. Filter pack #EF1 is not included in the average because it employed a quartz fiber prefilter and thus is not of the same configuration. The other filter packs, #JF1 and #NF1, were excluded because of systematic differences between day and nighttime sampling (#JF1), errors in chemical analysis (group N), and the large mean squared residual shown in Figure 7.

The denuder difference method is represented by those samplers using MgO coated denuders. Each of these samplers is preceded by a Teflon® cyclone or a Teflon®-coated cyclone to give a fine particle precut. Some of the DDM samplers use nylon filters; others use Teflon® and nylon filters in series. In all cases nitric acid is given by the difference between the denuded and undenuded sides of the sampler. The dichotomous samplers, which used nylon filters, and operated in parallel with a nylon filter behind a Teflon®-coated cyclone, also give nitric acid by the difference between denuded and undenuded sample streams. They are listed separately (Dichot) because the principal denuding action is from the aluminum oxide surfaces in the sampler inlet.

The annular denuder method average (ADM) is calculated from all three annular denuders, even though there are statistically significant differences among these samplers. The TFR and TDLAS averages are from duplicate instruments, which, as has been noted, show no significant differences. The FTIR data are represented by one instrument. The tungstic acid technique (TAT) is not included because of the large systematic differences between the two instruments.

Instantaneous Data

Instantaneous nitric acid values were reported by the two spectroscopic methods, the FTIR and TDLAS. These data, shown in Figure 10, represent five-minute averages from the two TDLAS instruments and are coincident with the most reliable FTIR data during the study period. Fifty-two data points from September 14 (n=34) and September 17 (n=18) are shown in the scatterplot, with the ratio of means TDLAS/FTIR equal to 0.84. At high nitric acid concentrations, the TDLAS data are even lower with respect to the FTIR.

Direct Period Comparison

Data from one sampling period (1200-1600 on September 14) permit direct comparison between the FTIR and the non-continuous sampling methods. The corresponding ratios to the reported FTIR period average of 605 neq/m3 are 1.32, 1.03, 0.95, and 0.84 for the FP, DDM, ADM and TDLAS, respectively. The closest agreement for this period is the DDM value of 626 neq/m^3, which is within 3% of the FTIR value. The FP are higher than the FTIR while the TDLAS is lower. The mean of the FP, DDM, ADM and TDLAS methods is also 626 neq/m3.

Sufficient data were obtained from the FTIR to give hourly average values for two or three of the four hours in sampling intervals 0800-1200 and 1600-2000 PDT on September 14 and for 1200-1600 PDT on September 17, 1986. To obtain a four-hour average for the FTIR during these periods, the missing FTIR hourly data are estimated by dividing the corresponding TDLAS data by the previously calculated correction factor of 0.84. These values are within the lower and upper bounds for the average concentration calculated by respectively assigning zero and the FTIR detection limit of 160 neq/m^3 to the missing hourly averages. The upper limit average is expected to be closer to the true period average, especially for the two periods on September 14 when the general level of pollution was high. The averages calculated by scaling to the TDLAS data are close to the upper bound calculation of the average concentrations. Data from the FTIR, TDLAS, FP, DDM and ADM are compared for each of these four-hour sampling periods in Figure 11. Error bars show the $\pm$160 neq/m3 measurement error for the FTIR, as reported by Winer et al. (1986). For periods with missing data, the additional error is as much

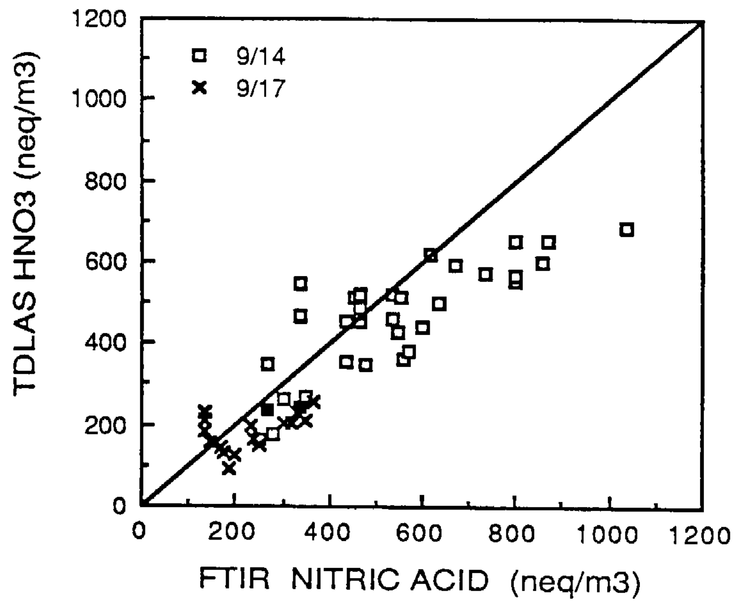

Figure 10. Comparison of 5 minute averages from the two tunable diode laser absorption spectrometers and the FTIR. The line of 1:1 correspondence is shown.

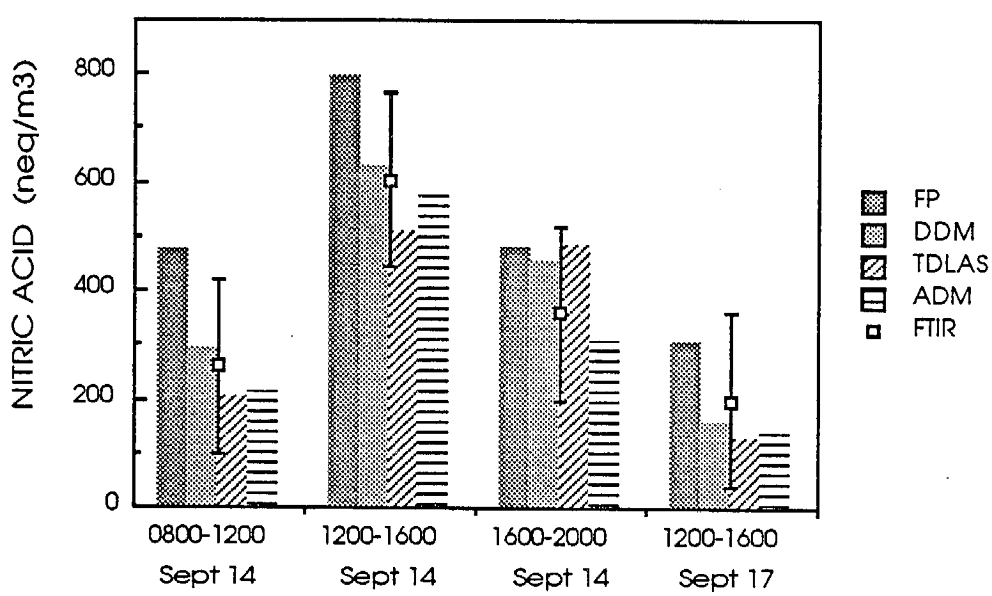

Figure 11. Methods comparison of nitric acid measurements for the high nitric acid periods of September 14 and the afternoon period of September 17. Error bars for the FTIR data correspond to ± 160 neq/m^3 ($\pm$4ppb).

as ±40neq/m^3. While the FTIR data averaged over the four periods are closest to the DDM method mean, all methods agree within their reported range of uncertainty.

Comparisons with the Mean of Methods

In order to compare reported nitric acid values by method throughout the study we calculated the mean of the methods, weighting the TDLAS, FP, ADM and DDM values equally. Parameters from linear regression of each method against the mean of methods are reported in Table 7. Data for the FP, TDLAS and DDM include 39 of the study periods, whereas the ADM data are for the 23 daytime periods when all three annular denuders were on the same schedule. Also shown is the ratio of each method to the mean of methods. This calculation shows FP values are 36% higher than the mean of the four methods, and that the TDLAS and ADM values are 13% and 21% less than the mean of methods. The ratio of the DDM to the mean of methods is 0.99. In the absence of a reference method for the entire study period, the DDM is chosen as an appropriate basis of comparison for the study, based on this and the previous period comparison with the FTIR.

Day - Night Comparison by Method

Since the DDM value agrees most closely with the mean of the methods and with the FTIR, we have plotted the ratio between each of the methods and the DDM (Figure 12). The data are shown for the daytime and nighttime periods of the study. Some of the data points represent composites of shorter sampling periods while others are from 10- and 12-hour samples. For easy comparison, the daytime data are given by open symbols, the nighttime data by solid symbols.

On the evening of September 16-17, the DDM mean appears to be low, as does the Dichot mean. During this period nitric acid concentration was low (DDM mean = 12 neq/m^3), and the ratio of HNO3 to fine particle nitrate was less than 0.06, which gives a large error in the denduder difference measurements.

Differences occur in the reported nitric acid levels between the TDLAS and other methods during the latter part of the study. On September 18, a period of intermittent drizzle, the TDLAS data are higher than values from the other methods. The TDLAS data do not exhibit as much diurnal variation as the other

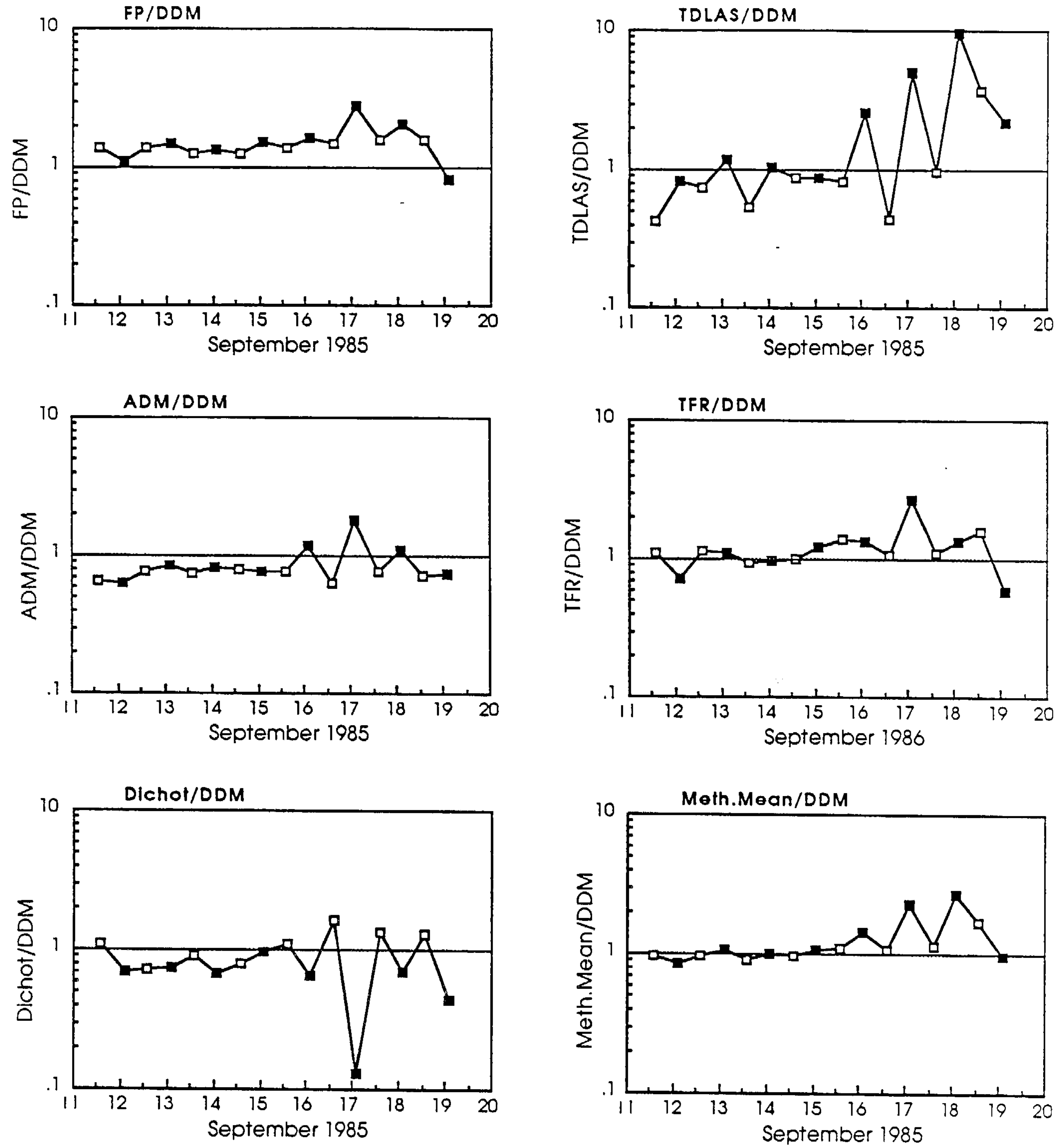

Figure 12. Ratio of the method means to the MgO denuder difference method mean (DDM) for the day and nighttime study periods. Methods shown are filter packs (FP), tunable diode laser absorption spectroscopy (TDLAS), annular denuders (ADM), transition flow reactors (TFR), and the dichotomous sampler denuder difference method (Dichot). Also shown is the mean of the methods with respect to the DDM.

methods, and thus with respect to the DDM reported concentrations are higher at night and lower during the day. On September 16, the TDLAS did not observe the afternoon nitric acid peak measured by other methods. Also, the TDLAS tended to be lower than the DDM at the beginning of the study period, and higher at the end.

Table 8 reports linear regression parameters for the TDLAS, FP, and ADM vs the DDM for the thirtynine 4- and 6-hour sampling periods (excluding period no. 64), and for the TFR and Dichot vs the DDM for the 10- and 12-hour sampling periods. Noteworthy are the day/night comparisons vs the DDM. The FP, TFR and ADM exhibit no diurnal variation relative to the DDM, while the TDLAS reports markedly higher values at night, averaging 65% higher than the DDM. The FP data are higher than the DDM for nighttime as well as daytime sampling. The work of Ellestad *et al.* (1986) and John *et al.* (1986) show that ammonium nitrate can lost from Teflon® filters during nighttime as well as daytime sampling.

Table 7. Linear regression parameters for nitric acid from each method versus the mean of methods.

x	y	n	Slope		Intercept (neq/m3)		r	$\Sigma y/\Sigma x$
Mean	TDLAS	39	0.75	+-0.09	17.	+-15.	0.94	0.87
Mean	FP	39	1.33	+-0.07	4.	+-14.	0.99	1.36
Mean	ADM	23	0.87	+-0.05	-15.	+-10.	0.99	0.79
Mean	DDM	39	1.07	+-0.04	-10.	+- 7.	0.99	0.99

n=number of data points
r=correlation coefficient
$\Sigma y/\Sigma x$=ratio of means
Period #64 excluded from analysis.

Table 8. Linear regression parameters for nitric acid from each method versus the mean denuder difference measurement.

x	y	Period	n	Slope		Intercept (neq/m3)		r	$\Sigma y/\Sigma x$
DDM	TDLAS	All	39	0.68	+-0.10	26.	+-18.	0.92	0.88
		Day	23	0.72	+-0.14	11.	+-28.	0.92	0.77
		Night	16	0.70	+-0.44	36.	+-25.	0.68	1.65
DDM	FP	All	39	1.23	+-0.08	18.	+-16.	0.98	1.37
		Day	23	1.20	+-0.12	30.	+-29.	0.98	1.35
		Night	16	1.35	+-0.18	5.	+-10.	0.98	1.49
DDM	ADM	All	39	0.79	+-0.05	-1.	+- 8.	0.99	0.78
		Day	23	0.81	+-0.07	-7.	+-14.	0.98	0.77
		Night *	16	0.73	+-0.11	5.	+- 5.	0.97	0.87
DDM	TFR	All	16	1.01	+-0.10	9.	+-16.	0.99	1.09
		Day	8	0.97	+-0.21	21.	+-40.	0.98	1.08
		Night	8	0.95	+-0.26	7.	+-11.	0.97	1.12
DDM	Dichot	All	16	0.86	+-0.12	8.	+-19.	0.97	0.93
		Day	8	0.76	+-0.22	40.	+-40.	0.96	0.97
		Night	8	0.74	+-0.18	-1.	+- 6.	0.97	0.71

n=number of data points
r=correlation coefficient
$\Sigma y/\Sigma x$=ratio of means

Period #64 excluded from analysis of 4- and 6-hour data.

* For this calculation, the single nighttime sample from sampler #EA1 was split into two samples of equal loadings, so as to match the sample frequency of the other data.

VI. References

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Appendix A. Description of Nitric Acid Measurement Methods

Filter Packs (FP): Filter packs were operated by several groups: the Canadian Atmospheric Environment Service (Anlauf and Wiebe, AES, Toronto, Canada), the California Institute of Technology (Cass and Solomon, CIT, Pasadena, CA), Daniel Grosjean and Associates (DGA, Ventura, CA), the Environmental Protection Agency (Martin and Pleasant, EPA, Research Triangle Park, NC) and National Oceanic and Atmospheric Administration (Womack, NOAA, Oak Ridge, TN).

Filter packs use two or more filters in series to remove both particles and gases. Usually a Teflon filter is used to remove particulate matter, followed by a Nylon filter to collect nitric acid. The exact configurations of the filter packs in this study are given in Table 1. The Canadian filter pack contains a citric acid impregnated filter following the Nylon filter to collect ammonia. The EPA filter pack has a K_2CO_3 impregnated filter for SO_2 collection. A filter pack operated by Brigham Young University consists of a quartz filter followed by a nylon filter. Most of the filter packs are open faced filters, and thus collected coarse as well as fine particle nitrate. The NOAA filter pack is preceded by a 0.5 m long horizontal elutriator to remove coarse particles.

Denuder Difference Methods (DDM): Denuder difference method samplers were operated by the Air Resources Board (Horrocks, ARB, El Monte, CA), Brigham Young University (Eatough), California Institute of Technology (CIT, Pasadena, CA, Cass and Solomon), Ford Motor Co. (Pierson and Brachaczek, Dearborn, MI) and two groups (John and Appel) from the Air and Industrial Hygiene Laboratory of the California Department of Health Services (AIHL, Berkeley, CA).

The denuder difference method employs two parallel sampling trains. In one train air is drawn through a diffusion denuder to remove gaseous nitric acid, followed by a nylon filter or a Teflon-nylon filter pack. The second sampling train is identical to the first except that the denuder is not present. The first sampling train collects particulate nitrate, while the second train collects the sum of nitric acid and particulate nitrate. Nitric acid is obtained by difference. Most of the denuder difference systems used a Teflon® or Teflon®-coated cyclone to give a coarse particle precut, followed by a MgO coated denuder.

Two of the denuder difference samplers (John, AIHL) use a standard Sierra-Andersen dichotomous sampler for the denuded side of the sampling train. The dichotomous sampler filter holders are modified to carry Teflon® and nylon filters mounted in series. In one dichotomous sampler, the aluminum oxide in the inlet of the sampler was tested as a means of removing the nitric acid. A second dichotomous sampler had an annular diffusion denuder of aluminum oxide added to the inlet. Coarse and fine particle nitrate is collected in the dichotomous samplers. A Teflon®-coated cyclone sampler followed by Teflon® and nylon filters in series was used to measure the sum of fine particle nitrate and nitric acid.

Annular Denuder Method (ADM): Annular denuders were operated by three groups. One system consisting of two annular denuders was operated cooperatively by Research Triangle Institute (RTI, Sickles) and the Italian National Research Council (CNR, Allegrini). Another system of five sequential annular denuders was operated by the University of Calgary (Peake). One annular denuder was operated by Brigham Young University (Eatough).

The annular denuder consists of two concentric glass cylinders coated with a chemical appropriate for the retention of the reactive gaseous species of interest. Air passes through the annular space between the two glass cylinders in a laminar flow where reactive gases are collected by diffusion. This geometry is quite efficient; at equivalent tube lengths and outer tube diameters larger sampling rates can be used than with an open tube denuder. Particles are collected on filters downstream of the denuder.

Upstream of the annular denuders in the RTI/CNR samplers (#IA1 and #IA2) a shrouded Teflon® cyclone is used to give a 2.5 μm precut, and a Teflon manifold is used to split the air flow into each of two identical sampling trains. In each train air passes in series through two 22 cm long Na_2CO_3 coated annular denuders to collect nitric acid, nitrous acid and sulfur dioxide; one 13 cm long citric acid coated annular denuder to collect gaseous ammonia; one Teflon filter to collect fine particles, one nylon filter to collect volatilized nitrate; and one 13 cm citric acid coated annular denuder to collect volatilized ammonia.

There are two major differences between the annular denuder and the denuder difference method. First, with the annular denuder the gaseous species depositing in the denuder are analyzed directly. Thus nitric acid is obtained directly rather than by the difference of two measurements. The second difference is in the design of the denuder itself.

Transition Flow Reactor (TFR): This system was operated by two groups, EPA (Ellestad) and Battelle Columbus (Spicer). It was designed for network monitoring of the concentrations of acidic species, which can be used to estimate dry deposition fluxes. Air is drawn through a cyclone which removes particles larger than 2 μm diameter. At the cyclone's outlet, a vortex-removing tee disrupts the spin circulation set up by the cyclone and splits the flow into two 16.1 std L/min sample streams. Identical sampling trains are used for each of these sample flows. Each 16.1 std L/min sample enters a tube lined with 3.2 cm long Nylon and Nafion strips, which remove 9% of the gaseous nitric acid and 17% of the gaseous ammonia, respectively. Each sample then passes through a Teflon® filter to remove particles, a Nylon filter to collect the remaining gaseous nitric acid and any nitric acid gas from the decomposition of ammonium nitrate on the Teflon filter, and an oxalic acid impregnated filter to collect the remaining ammonia and any ammonia from the decomposition of ammonium nitrate on the Teflon ® filter. The flow stream is then split again. The first stream of 1.78 L/min flow goes through triethanolamine impregnated glass fiber filters for SO_2 and NO_2 collection and then passes through a flow controller to the pump. The second stream of 16 L/min passes through another mass flow controller to the same pump. All surfaces in contact with the sample stream are Teflon®.

Tunable Diode Laser Absorption Spectrometer (TDLAS): Tunable diode laser absorption spectrometers were operated by the Canadian Atmospheric Environment Service (Anlauf and Wiebe, AES, Toronto, Canada) and by Unisearch Associates (Mackay, Toronto, Canada). The instruments were essentially identical in design, both having been manufactured by Unisearch.

The TDLAS detects species such as nitric acid by infrared absorption in the 2 to 15 μm region, as described by Hastie *et al.* (1984) Measurements are made utilizing a long-path White cell, operating at approximately 25 torr. The White cell is a Teflon lined 1 m long, 11.5 cm ID pyrex tube. The low pressure is needed to prevent collisional broadening of spectral lines. Ambient air is introduced into the White cell through a 6 mm OD, 0.75 mm wall Teflon tube. Particles are removed by an in-line 0.2µm pore Teflon filter located at the entrance of the sampling line. A Teflon needle valve located downstream of the filter maintains the flow at about 5 L/min.

Tungstic Acid Technique (TAT): Two TAT systems were used in the study, one operated by the University of South Florida (Braman), and one operated by the Air and Industrial Hygiene Laboratory (Appel). These are semi-continuous methods which utilize the fact that nitric acid is readily deposited on room temperature tungstic acid-coated surfaces, but at higher temperatures desorbs as an oxide of nitrogen. Air is sampled through a tungstic acid coated preconcentrator tube at 1 L/min for about 10 minutes. Then helium carrier gas is introduced while the tube is heated. The deposited nitric acid desorbs as NO or NO_2 and the deposited ammonia desorbs unchanged. Ammonia is then trapped in a WO_x coated transfer tube, while the NO_x passes through to a gold catalyst, where it is converted to NO and detected with a commercial chemiluminescent analyzer. The transfer tube is then heated to desorb the ammonia, which is then oxidized to NO over the gold catalyst and measured by chemiluminescence.

Fourier Transform Infrared Spectroscopy (FTIR): The FTIR was operated by the University of California, Riverside (Winer and Tuazon), to give measurements of ambient nitric acid and ammonia (Tauzon *et al.,* 1981). In this experiment the FTIR used an open multiple reflection optical system with a 25 m basepath, operated at a total pathlength of 1150m. The entire system is elevated 2.5 m above the ground using large cement headers. Spectra are recorded with a liquid nitrogen cooled HgCdTe detector at 0.13 cm^{-1} spectral resolution using 5-minute averaging times. Four to five spectra are obtained per hour. The detection limit for nitric acid in this study was 4 ppb (160 neq/m^3).

Luminol HNO_3: This is a semi-continuous method operated by the University of Denver (Stedman, Denver, CO). The total ambient NO_x and nitric acid is measured by passing air through a bed of hot glass beads to a luminol NO_2 detector. The hot beads convert NO and HNO_3 to NO_2. The inlet airstream is alternately chopped with a Teflon filter and a Teflon-Nylon filter pack. Nitric acid is obtained by the difference between the two signals.

Appendix B. Data Listing

The period averaged concentrations for the nitrogenous and sulfur species are given in the successive tables. Samplers are listed by ID numbers, as given in Table 1. Samples are listed by period number, of which the first digit corresponds to the date, and the second digit to the time of day, as follows:

Prd No.	Starting Date	Sampling Time (PDT)	Duration (hours)
12	Sept. 11, 1985	0800-1200	4
13	Sept. 11, 1985	1200-1600	4
14	Sept. 11, 1985	1600-2000	4
15	Sept. 11, 1985	2000-0600	4
16	Sept. 11, 1985	0800-2000	12
17	Sept. 11, 1985	2000-0600	10
18	Sept. 11, 1985	0800-0600	22
21	Sept. 12, 1985	0000-0600	6
22	Sept. 12, 1985	0800-1200	4
23	Sept. 12, 1985	1200-1600	4
24	Sept. 12, 1985	1600-2000	4
25	Sept. 12, 1985	2000-0600	4
26	Sept. 12, 1985	0800-2000	12
27	Sept. 12, 1985	2000-0600	10
28	Sept. 12, 1985	0800-0600	22
31	Sept. 13, 1985	0000-0600	6
32	Sept. 13, 1985	0800-1200	4
33	Sept. 13, 1985	1200-1600	4
34	Sept. 13, 1985	1600-2000	4
35	Sept. 13, 1985	2000-0600	4
36	Sept. 13, 1985	0800-2000	12
37	Sept. 13, 1985	2000-0600	10
38	Sept. 13, 1985	0800-0600	22
.	.	.	
.	.	.	
.	.	.	
81	Sept. 18, 1985	0000-0600	6
82	Sept. 18, 1985	0800-1200	4
83	Sept. 18, 1985	1200-1600	4
84	Sept. 18, 1985	1600-2000	4
85	Sept. 18, 1985	2000-0600	4
86	Sept. 18, 1985	0800-2000	12
87	Sept. 18, 1985	2000-0600	10
88	Sept. 18, 1985	0800-0600	22
91	Sept. 19, 1985	0000-0600	6

NITRIC ACID (nanomoles/m3)

PRD	AC1	AD1	BD1	BD3	CC1	CF1	CF3	CF2	DE1	DE2	EA1	ED1	EF1	FD1	GD1	GD2	GD3	GF1
12	.	16.	.	.	7.	31.	.	34.	.	.	35.	24.	19.	15.	26.	25.	20.	15.
13	.	86.	.	.	35.	150.	.	144.	.	.	114.	32.	44.	87.	85.	89.	86.	83.
14	.	55.	.	.	31.	97.	.	100.	.	.	92.	31.	18.	75.	79.	73.	63.	71.
15	.	19.	.	.	3.	17.	.	20.	.	.	.	.	.	13.	22.	23.	10.	23.
21	.	16.	.	.	4.	12.	.	13.	.	.	.	.	.	10.	.	.	.	.
22	.	113.	.	.	85.	204.	.	203.	.	.	75.	213.	220.	69.	132.	.	113.	134.
23	.	139.	.	.	119.	239.	.	261.	.	.	103.	129.	198.	242.	156.	.	134.	.
24	.	364.	.	.	350.	474.	.	487.	.	.	471.	185.	315.	341.	306.	.	441.	468.
25	.	132.	.	.	110.	151.	.	153.	.	.	.	.	.	20.	142.	.	84.	147.
31	.	55.	.	.	55.	70.	.	63.	.	.	.	.	.	76.	52.	.	35.	71.
32	.	.	.	.	72.	264.	.	215.A	.	.	202.	206.	182.	134.	199.	.	132.	179.
33	.	439.	.	.	266.	549.	.	466.A	.	.	299.	358.	406.	446.	442.	.	435.	485.
34	.	497.	.	.	274.	643.	.	.	.	.	506.	460.	516.	566.	668.	.	425.	606.
35	.	165.	.	.	130.	188.	.	166.A	.	.	.	.	.	128.	156.	.	149.	180.
41	.	113.	.	.	85.	97.	.	93.A	.	.	.	.	.	78.	87.	.	64.	115.
42	.	408.	.	.	180.	473.	.	.	.	.	231.	129.	288.	198.	367.	.	331.	492.
43	429.	765.	.	.	503.	795.	.	729.A	.	.	684.	488.	670.	613.	578.	.	.	788.
44	596.	503.	.	.	447.	502.	.	448.A	.	.	423.	405.	402.	431.	444.	.	469.	498.
45	391.	105.	.	.	59.	102.	.	71.A	.	.	.	.	.	55.	74.	.	73.	104.
51	280.	48.	.	.	16.	59.	.	40.A	.	.	.	.	.	36.	52.	.	47.	74.
52	286.	260.	.	.	138.	304.	.	240.A	.	.	203.	430.	231.	113.	187.	.	185.	302.
53	362.	295.	.	.	247.	503.	.	367.A	.	.	298.	240.	410.	332.	327.	.	465.	406.
54	261.	155.	.	.	154.	221.	.	208.A	.	.	181.	147.	121.	197.	206.	.	218.	224.
55	123.	11.	.	.	55.	52.	.	50.A	.	.	.	.	.	43.	16.	.	34.	59.
61	93.	16.	.	.	55.	24.	.	24.A	.	.	.	.	.	40.	25.	.	15.	42.
62	47.	95.	.	.	26.	47.	.	60.A	.	.	21.	287.	315.	33.	110.	.	.	67.
63	123.	147.	.	.	42.	307.	.	234.A	.	.	149.	31.	167.	120.	175.	.	.	270.
64	103.	89.	.	.	.	87.	.	85.A	.	.	71.	.	39.	112.	83.	.	.	116.
65	52.	26.	.	.	30.	22.	.	25.A	.	.	.	.	.	2.	10.	.	.	21.
71	.	31.	.	.	89.	47.	.	43.A	.	.	.	.	.	7.	2.	.	.	42.
72	61.	98.	.	.	30.	48.	44.	50.A	.	.	15.	58.	26.	0.	0.	.	.	62.
73	121.	163.	.	.	140.	344.	317.	274.A	.	.	184.	110.	250.	134.	159.	.	.	273.
74	.	89.	.	.	119.	137.	134.	144.A	.	.	86.	.	78.	73.	111.	.	.	105.
75	103.	13.	.	.	65.	27.	.	30.A	.	.	.	.	.	10.	2.	.	.	6.
81	.	10.	.	.	81.	18.	.	18.A	.	.	.	.	.	8.	10.	.	.	24.
82	96.	11.	.	.	55.	25.	.	22.A	.	.	8.	33.	11.	7.	0.	.	.	28.
83	119.	31.	.	.	119.	55.	.	43.A	.	.	.	26.	22.	26.	.	.	.	52.
84	161.	19.	.	.	82.	23.	.	19.A	.	.	28.	20.	10.	30.	5.	.	.	13.
85	166.	5.	.	.	43.	12.	.	8.A	.	.	.	.	.	0.	16.	.	.	9.
91	166.	5.	.	.	4.	.	4.	6.A	.	.	.	.	.	6.	32.	.	.	12.
16	.	52.A	64.	64.	24.A	93.A	.	93.A	61.	.	53.	29.A	27.A	59.A	63.A	62.A	56.A	56.A
17	.	17.A	8.	12.	3.A	14.A	.	16.A	8.	.	10.	.	8.	11.A	.	.	.	.
26	.	205.A	150.	150.	185.A	306.A	.	317.A	.	.	211.	176.A	244.A	217.A	198.A	.	229.A	.
27	.	86.A	47.	51.	77.A	102.A	.	99.A	.	.	75.	158.	55.	54.A	88.A	.	55.A	101.A
36	.	.	330.	328.	204.A	485.A	.	.	.	.	415.	341.A	368.A	382.A	436.A	.	330.A	424.A
37	.	134.A	68.	73.	103.A	133.A	.	.	101.	.	100.	64.	75.	98.A	114.A	.	98.A	141.A
46	.	559.A	382.	355.	377.A	590.A	.	.	463.	.	550.	377.	433.	414.A	463.A	.	.	593.A
47	325.A	71.A	49.	51.	33.A	76.A	.	.	67.	.	25.	41.	32.	44.A	61.A	.	58.A	86.A
56	303.A	237.A	267.	244.	180.A	343.A	.	.	399.	.	191.	272.A	254.A	214.A	240.A	.	289.A	311.A
57	105.A	14.A	13.	17.	55.A	35.A	.	.	42.	.	24.	16.	15.	41.A	22.A	.	23.A	49.A
66	91.A	110.A	161.	151.	36.A	147.A	.	.	101.	.	70.	.	173.A	88.A	123.A	.	.	151.A
67	.	29.A	0.	5.	65.A	37.A	.	.	41.	.	25.	.	19.	5.A	6.A	.	.	34.A
76	.	117.A	136.	138.	96.A	176.A	165.A	.	101.	.	95.	.	118.A	69.A	90.A	.	.	147.A
77	.	11.A	4.	10.	75.A	22.A	.	.	11.	.	13.	.	7.	9.A	7.A	.	.	17.A
86	126.A	20.A	23.	24.	85.A	34.A	.	.	36.	.	22.	27.A	14.A	21.A	.	.	.	31.A
87	166.A	5.A	4.	4.	19.A	.	.	.	13.	.	13.	7.	7.	4.A	25.A	.	.	11.A
18	.	36.A	38.A	40.A	15.A	57.A	.	58.A	41.A	.	33.A	.	18.A	37.A	.	.	.	.
28	.	151.A	103.A	105.A	136.A	213.A	.	218.A	.	.	149.A	167.A	158.A	143.A	148.A	158.	150.A	.
38	.	.	211.A	212.A	158.A	325.A	.	.	.	488.	272.A	215.A	235.A	253.A	290.A	231.	225.A	295.A
48	.	337.A	231.A	217.A	221.A	357.A	.	.	283.A	.	311.A	224.A	251.A	246.A	280.A	292.	.	362.A
58	213.A	136.A	152.A	141.A	123.A	203.A	.	.	237.A	157.	115.A	156.A	145.A	136.A	141.A	166.	168.A	192.A
68	.	73.A	88.A	85.A	48.A	97.A	.	.	74.A	56.	50.A	.	103.A	51.A	69.A	68.	77.	98.A
78	.	69.A	76.A	80.A	86.A	106.A	.	.	60.A	72.	58.A	.	68.A	42.A	52.A	68.	82.	88.A
88	144.A	13.A	15.A	15.A	55.A	.	.	.	25.A	24.	18.A	18.A	11.A	13.A	.	19.	19.	22.A

NITRIC ACID (nanomoles/m3)

Prd	GF2	GF3	HE1	HE2	IA1	IA2	JF1	JF2	KD1	KD2	MF1	NF1	NF2	PC1	QA1	RC1	SC1	TC1
12	42.	26.	.	.	4.	3.	.	.	15.	.	.	.	.	.	3.	.	.	12.
13	102.	104.	.	.	14.	16.	.	.	89.	.	139.	.	.	.	7.	.	113.	10.
14	71.	82.	.	.	29.	16.	.	.	62.	.	108.	.	.	32.	37.	.	82.	33.
15	12.	12.	.	.	14.	10.	.	.	18.	.	18.	.	.	28.	8.	.	111.	38.
21	.	.	.	.	3.	3.	.	.	5.	.	.	.	.	9.	.	.	92.	21.
22	.	165.	.	.	37.	38.	.	.	88.	.	79.	.	.	57.	130.	.	155.	59.
23	.	194.	.	.	69.	44.	.	.	107.	.	.	.	.	76.	92.	.	129.	71.
24	.	441.	.	.	177.	168.	.	.	348.	.	482.	.	.	239.	290.	.	283.	153.
25	.	122.	.	.	68.	40.	.	.	65.	.	129.	.	.	.	108.	.	249.	120.
31	.	46.	.	.	21.	24.	.	.	32.	.	66.	.	.	59.	25.	.	70.	80.
32	.	196.	.	.	53.	51.	.	.	111.	.	.	.	.	50.	132.	.	.	96.
33	.	500.	.	.	268.	191.	.	.	367.	.	.	.	.	228.	330.	.	.	186.
34	.	586.	.	.	321.	.	.	.	466.	.	.	.	.	262.	371.	.	.	208.
35	.	187.	.	.	74.	.	.	.	122.	.	.	.	.	178.	127.	.	.	168.
41	.	113.	.	.	42.	.	.	.	49.	.	.	.	.	.	77.	.	.	127.
42	.	452.	.	.	182.	.	.	.	218.	.	.	.	.	227.	226.	.	.	139.
43	.	800.	.	.	511.	.	.	.	547.	.	.	.	.	514.	524.	605.	305.	191.
44	.	412.	.	.	168.	.	.	.	423.	.	.	.	.	518.	325.	320.	243.	188.
45	.	114.	.	.	58.	.	.	.	57.	.	.	.	.	53.	72.	.	.	145.
51	.	75.	.	.	31.	.	.	.	19.	.	.	.	.	63.	40.	.	.	93.
52	.	283.	.	.	88.	.	.	.	139.	.	.	.	.	132.	129.	.	203.	.
53	.	358.	.	.	204.	.	.	.	327.	.	.	.	.	255.	255.	.	149.	75.
54	.	203.	.	.	120.	.	.	.	183.	.	.	.	.	214.	154.	.	98.	52.
55	.	39.	.	.	18.	.	.	.	29.	.	.	.	.	82.	49.	.	.	64.
61	.	38.	.	.	12.	.	.	.	12.	.	.	.	.	55.	40.	.	.	59.
62	.	55.	.	.	22.	.	.	.	-12.	.	.	.	.	50.	27.	.	203.	77.
63	.	178.	.	.	79.	.	.	.	120.	.	.	.	.	39.	109.	.	80.	75.
64	.	100.	.	.	.	9.	.	.	72.	.	.	.	.	.	60.	.	45.	57.
65	.	20.	.	.	.	.	.	.	10.	.	.	.	.	.	19.	.	90.	39.
71	.	35.	.	.	.	.	.	.	7.	.	.	.	.	99.	19.	.	33.	36.
72	.	15.	.	.	.	12.	.	.	-3.	.	.	.	.	60.	23.	.	51.	51.
73	.	272.	.	.	.	115.	.	.	178.	.	.	.	.	117.	120.	200.	50.	71.
74	.	129.	.	.	.	102.	.	.	88.	.	.	.	.	134.	59.	.	23.	60.
75	.	7.	.	.	.	8.	.	.	13.	.	.	.	.	107.	15.	.	15.	22.
81	.	18.	.	.	.	3.	.	.	6.	.	.	.	.	94.	8.	.	19.	23.
82	.	25.	.	.	4.	5.	.	.	5.	.	26.	.	.	39.	11.	.	39.	26.
83	.	7.	.	.	10.	11.	.	.	24.	.	.	.	.	77.	29.	.	39.	20.
84	.	8.	.	.	9.	7.	.	.	13.	.	29.	.	.	36.	18.	.	38.	19.
85	.	7.	.	.	5.	6.	.	.	3.	.	60.	.	.	.	6.	.	22.	13.
91	.	10.	.	.	4.	3.	.	.	5.	.	8.	.	.	18.	7.	.	11.	8.
16	71.A	71.A	65.	.	16.A	12.A	31.	0.	56.A	.	.	24.	.	32.A	16.A	.	.	21.A
17	.	.	12.	.	8.A	6.A	0.	0.	10.A	.	.	9.	.	17.A	.	.	100.A	27.A
26	.	266.A	234.	.	94.A	83.A	317.	305.	181.A	.	.	238.	.	101.A	171.A	.	189.A	91.A
27	.	77.A	73.	.	40.A	30.A	44.	51.	45.A	.	91.A	67.	.	59.A	58.A	.	142.A	89.A
36	.	427.A	341.	.	214.A	.	523.	501.	315.A	.	.	318.	.	180.A	278.A	.	.	162.A
37	.	142.A	96.	.	55.A	50.	86.	84.	78.A	.	.	56.	.	178.A	97.A	.	.	142.A
46	.	555.A	446.	.	287.A	10.	781.	762.	396.A	.	.	333.	.	420.A	358.A	.	.	169.A
47	.	91.A	62.	.	42.A	27.	11.	28.	34.A	.	.	9.	.	59.A	53.A	.	.	117.A
56	.	282.A	239.	.	137.A	165.	499.	479.	216.A	.	.	238.	.	200.A	179.A	.	150.A	64.A
57	.	38.A	23.	.	14.A	11.	0.	0.	19.A	.	.	29.	.	66.A	44.A	.	.	61.A
66	.	111.A	104.	.	.	.	149.	201.	60.A	.	.	149.	.	48.A	65.A	.	110.A	70.A
67	.	29.A	23.	.	.	7.	0.	0.	8.A	.	.	57.	.	99.A	19.A	.	56.A	38.A
76	.	139.A	121.	.	56.	76.A	151.	175.	88.A	.	.	29.	.	104.A	67.A	.	42.A	61.A
77	.	14.A	14.	.	5.	5.A	0.	0.	9.A	.	.	19.	.	99.A	11.A	.	17.A	23.A
86	.	13.A	23.	.	8.A	7.A	0.	0.	14.A	.	.	16.	.	51.A	19.A	.	39.A	21.A
87	.	9.A	0.	.	5.A	4.A	0.	0.	4.A	.	29.A	5.	.	33.A	7.A	.	15.A	10.A
18	.	.	41.A	39.	12.A	9.A	17.A	0.A	35.A	33.	.	17.A	30.	21.A	.	.	.	24.A
28	216.	180.A	160.A	136.	70.A	59.A	193.A	190.A	119.A	87.	.	160.A	.	99.A	120.A	.	167.A	90.A
38	353.	298.A	230.A	190.	142.A	.	325.A	312.A	207.A	145.	.	199.A	.	179.A	196.A	.	.	151.A
48	416.	344.A	271.A	240.	175.A	18.A	431.A	428.A	231.A	144.	.	186.A	.	256.A	219.A	.	.	142.A
58	.	171.A	141.A	121.	81.A	95.A	272.A	261.A	127.A	105.	.	143.A	.	139.A	118.A	.	.	62.A
68	115.	73.A	68.A	59.	.	.	81.A	110.A	36.A	32.	.	107.A	.	67.A	44.A	.	85.A	53.A
78	115.	82.A	72.A	60.	33.A	44.A	82.A	96.A	52.A	34.	.	24.A	.	102.A	41.A	.	31.A	45.A
88	27.	11.A	13.A	14.	6.A	6.A	0.A	0.A	10.A	11.	.	11.A	3.	44.A	14.A	.	28.A	16.A

FINE NITRATE (nanomoles/m3)

Prd	AD1	BD1	BD3	DE1	DE2	EA1	ED1	EF1	FD1	GD1	GD2	GD3	GF1
12	26.	.	.	.	.	30.	30.	35.	15.	25.	19.	28.	9.
13	37.	.	.	.	.	49.	49.	37.	27.	42.	31.	37.	6.
14	29.	.	.	.	.	22.	22.	35.	26.	31.	33.	30.	12.
15	11.	.	.	.	.	.	.	.	11.	11.	10.	16.	9.
21	30.	.	.	.	.	.	.	.	18.	.	.	.	.
22	117.	.	.	.	.	120.	120.	113.	115.	96.	.	119.	24.
23	197.	.	.	.	.	233.	233.	163.	164.	198.	.	212.	50.
24	237.	.	.	.	.	285.	285.	156.	233.	241.	.	204.	73.
25	142.	.	.	.	.	.	.	.	166.	78.	.	119.	71.
31	115.	.	.	.	.	.	.	.	62.	116.	.	121.	89.
32	.	.	.	.	.	128.	128.	152.	172.	171.	.	204.	86.
33	160.	.	.	.	.	184.	184.	136.	129.	157.	.	165.	59.
34	145.	.	.	.	.	159.	159.	103.	119.	28.	.	148.	51.
35	75.	.	.	.	.	.	.	.	77.	80.	.	87.	39.
41	152.	.	.	.	.	.	.	.	147.	186.	.	189.	147.
42	497.	.	.	.	.	487.	487.	328.	579.	582.	.	598.	291.
43	267.	.	.	.	.	326.	326.	145.	270.	292.	.	298.	47.
44	69.	.	.	.	.	85.	85.	88.	80.	85.	.	83.	25.
45	30.	.	.	.	.	.	.	.	54.	41.	.	43.	21.
51	56.	.	.	.	.	.	.	.	52.	67.	.	67.	46.
52	484.	.	.	.	.	375.	375.	574.	449.	515.	.	512.	204.
53	169.	.	.	.	.	229.	229.	59.	202.	182.	.	57.	12.
54	57.	.	.	.	.	57.	57.	84.	40.	65.	.	61.	23.
55	87.	.	.	.	.	.	.	.	71.	24.	.	61.	54.
61	93.	.	.	.	.	.	.	.	45.	95.	.	97.	77.
62	362.	.	.	.	.	373.	373.	345.	406.	392.	.	.	367.
63	206.	.	.	.	.	231.	231.	95.	205.	201.	.	.	22.
64	52.	.	.	.	.	66.	66.	.	23.	87.	.	.	43.
65	69.	.	.	.	.	.	.	.	68.	81.	.	.	66.
71	153.	.	.	.	.	.	.	.	162.	175.	.	.	155.
72	385.	.	.	.	.	379.	379.	411.	413.	464.	.	.	403.
73	233.	.	.	.	.	263.	263.	124.	219.	238.	.	.	17.
74	77.	.	.	.	.	179.	179.	97.	83.	97.	.	.	33.
75	45.	.	.	.	.	.	.	.	55.	52.	.	.	32.
81	38.	.	.	.	.	.	.	.	48.	40.	.	.	28.
82	34.	.	.	.	.	38.	38.	60.	44.	38.	.	.	24.
83	35.	.	.	.	.	63.	63.	67.	27.	31.	.	.	17.
84	13.	.	.	.	.	25.	25.	36.	13.	12.	.	.	11.
85	26.	.	.	.	.	.	.	.	20.	22.	.	.	25.
91	22.	.	.	.	.	.	.	.	29.	15.	.	.	24.
16	31.A	32.	32.	32.	.	35.	35.	36.A	23.A	32.A	28.A	32.A	9.A
17	22.A	34.	30.	20.	.	19.	19.	.	15.A	.	.	.	.
26	184.A	165.	165.	.	.	117.	117.	144.A	171.A	179.A	.	179.A	49.A
27	126.A	118.	114.	.	.	16.	16.	119.	104.A	101.A	.	120.A	82.A
36	.	144.	147.	.	.	160.	160.	130.A	140.A	119.A	.	172.A	65.A
37	121.A	126.	120.	148.	.	131.	131.	120.	119.A	144.A	.	148.A	104.A
46	278.A	281.	307.	342.	.	69.	69.	13.	310.A	320.A	.	327.A	121.A
47	46.A	59.	57.	26.	.	50.	50.	58.	53.A	57.A	.	57.A	36.A
56	237.A	256.	278.	170.	.	254.	254.	239.A	230.A	254.A	.	210.A	80.A
57	91.A	97.	93.	19.	.	70.	71.	71.	55.A	67.A	.	83.A	68.A
66	207.A	190.	199.	214.	.	216.	216.	.	211.A	227.A	.	.	144.A
67	120.A	132.	128.	35.	.	116.	116.	86.	124.A	137.A	.	.	119.A
76	232.A	226.	225.	186.	.	234.	234.	211.A	238.A	266.A	.	.	151.A
77	40.A	56.	50.	32.	.	39.	39.	13.	51.A	45.A	.	.	30.A
86	27.A	44.	44.	26.	.	34.	34.	54.A	28.A	27.A	.	.	17.A
87	24.A	34.	33.	14.	.	30.	30.	31.	25.A	18.A	.	.	25.A
18	27.A	33.A	31.A	26.A	.	27.A	27.A	.	19.A	.	.	.	.
28	157.A	143.A	142.A	.	.	72.A	72.A	133.A	140.A	143.A	171.	152.A	64.A
38	.	136.A	135.A	.	0.	147.A	147.A	126.A	131.A	130.A	152.	161.A	83.A
48	172.A	180.A	194.A	199.A	.	60.A	60.A	33.A	193.A	200.A	192.	204.A	82.A
58	170.A	183.A	194.A	102.A	146.	170.A	171.A	163.A	151.A	169.A	181.	152.A	74.A
68	167.A	164.A	167.A	132.A	110.	170.A	170.A	.	172.A	186.A	186.	186.	133.A
78	145.A	149.A	145.A	116.A	158.	145.A	145.A	121.A	153.A	166.A	168.	158.	96.A
88	26.A	40.A	39.A	21.A	15.	32.A	32.A	44.A	27.A	23.A	31.	29.	21.A

FINE NITRATE (nanomoles/m3)

Prd	GF2	GF3	HE1	HE2	IA1	IA2	JF1	JF2	KD1	KD2	NF1	NF2	QA1	TC1
12	8.	7.	.	.	4.	5.	.	.	24.	.	.	.	12.	361.
13	6.	5.	.	.	6.	5.	.	.	36.	.	.	.	31.	11.
14	9.	8.	.	.	7.	10.	.	.	35.	.	.	.	20.	215.
15	9.	10.	.	.	9.	11.	.	.	11.	.	.	.	17.	45.
21	.	.	.	.	8.	14.	.	.	29.	.	.	.	.	42.
22	.	16.	.	.	39.	37.	.	.	109.	.	.	.	68.	130.
23	.	54.	.	.	84.	64.	.	.	213.	.	.	.	108.	191.
24	.	75.	.	.	91.	86.	.	.	254.	.	.	.	127.	317.
25	.	68.	.	.	29.	40.	.	.	141.	.	.	.	80.	163.
31	.	86.	.	.	46.	43.	.	.	123.	.	.	.	52.	129.
32	.	102.	.	.	83.	66.	.	.	202.	.	.	.	126.	198.
33	.	63.	.	.	54.	48.	.	.	163.	.	.	.	108.	233.
34	.	76.	.	.	45.	.	.	.	150.	.	.	.	69.	290.
35	.	41.	.	.	0.	.	.	.	85.	.	.	.	54.	181.
41	.	142.	.	.	59.	.	.	.	171.	.	.	.	60.	205.
42	.	341.	.	.	301.	.	.	.	520.	.	.	.	159.	300.
43	.	57.	.	.	122.	.	.	.	278.	.	.	.	139.	250.
44	.	19.	.	.	18.	.	.	.	75.	.	.	.	53.	101.
45	.	23.	.	.	10.	.	.	.	33.	.	.	.	35.	91.
51	.	45.	.	.	26.	.	.	.	61.	.	.	.	42.	91.
52	.	258.	.	.	234.	.	.	.	527.	.	.	.	285.	.
53	.	10.	.	.	57.	.	.	.	194.	.	.	.	139.	52.
54	.	29.	.	.	14.	.	.	.	50.	.	.	.	67.	28.
55	.	52.	.	.	23.	.	.	.	59.	.	.	.	56.	63.
61	.	83.	.	.	31.	.	.	.	84.	.	.	.	81.	86.
62	.	.	.	.	153.	.	.	.	408.	.	.	.	308.	241.
63	.	.	.	.	60.	.	.	.	194.	.	.	.	88.	142.
64	.	.	.	.	.	43.	.	.	45.	.	.	.	70.	56.
65	.	.	.	.	.	.	.	.	65.	.	.	.	66.	36.
71	.	.	.	.	.	.	.	.	124.	.	.	.	78.	28.
72	.	.	.	.	.	200.	.	.	445.	.	.	.	323.	60.
73	.	.	.	.	.	103.	.	.	238.	.	.	.	189.	54.
74	.	.	.	.	.	37.	.	.	72.	.	.	.	90.	13.
75	.	.	.	.	.	11.	.	.	41.	.	.	.	47.	14.
81	.	.	.	.	.	12.	.	.	36.	.	.	.	35.	10.
82	.	.	.	.	8.	12.	.	.	35.	.	.	.	28.	9.
83	.	.	.	.	11.	7.	.	.	40.	.	.	.	15.	11.
84	.	.	.	.	1.	7.	.	.	17.	.	.	.	23.	6.
85	.	.	.	.	10.	8.	.	.	30.	.	.	.	21.	4.
91	.	.	.	.	15.	13.	.	.	24.	.	.	.	23.	4.
16	8.A	7.A	35.	.	6.A	7.A	3.	8.	32.A	.	40.	.	21.A	273.
17	.	.	14.	.	9.A	13.A	0.	7.	21.A	.	57.	.	.	43.
26	.	48.A	203.	.	71.A	62.A	120.	194.	192.A	.	78.	.	101.A	210.
27	.	79.A	112.	.	39.A	42.A	122.	101.	130.A	.	67.	.	63.A	137.
36	.	80.A	230.	.	61.A	.	77.	48.	172.A	.	56.	.	101.A	241.
37	.	102.A	140.	.	35.A	38.	134.	115.	137.A	.	56.	.	58.A	197.
46	.	139.A	365.	.	147.A	145.	117.	85.	291.A	.	64.	.	117.A	210.
47	.	36.A	42.	.	19.A	10.	16.	102.	50.A	.	57.	.	39.A	91.
56	.	99.A	243.	.	102.A	106.	88.	63.	257.A	.	106.	.	164.A	40.
57	.	71.A	55.	.	28.A	19.	211.	101.	74.A	.	159.	.	71.A	77.
66	.	.	175.	.	.	.	221.	102.	216.A	.	.	.	155.A	139.
67	.	.	93.	.	.	65.	233.	58.	100.A	.	175.	.	73.A	31.
76	.	.	220.	.	84.	113.A	255.	85.	252.A	.	508.	.	201.A	43.
77	.	.	28.	.	19.	12.A	12.	126.	38.A	.	84.	.	40.A	11.
86	.	.	21.	.	7.A	9.A	5.	77.	31.A	.	40.	.	22.A	8.
87	.	.	26.	.	13.A	11.A	0.	115.	27.A	.	29.	.	22.A	4.
18	.	.	25.A	28.	7.A	9.A	2.A	8.A	27.A	28.	48.A	49.	.	175.
28	71.	62.A	162.A	177.	57.A	53.A	121.A	152.A	164.A	171.	73.A	.	84.A	175.
38	71.	90.A	189.A	222.	49.A	.	103.A	78.A	156.A	161.	56.A	.	81.A	217.
48	40.	92.A	218.A	243.	89.A	84.A	71.A	93.A	181.A	192.	61.A	.	82.A	148.
58	42.	86.A	158.A	176.	68.A	66.A	144.A	80.A	174.A	177.	130.A	.	122.A	65.
68	82.	81.	138.A	157.	.	.	227.A	82.A	163.A	171.	.	.	118.A	82.
78	57.	58.	132.A	149.	55.A	67.A	144.A	104.A	154.A	153.	315.A	.	128.A	30.
88	23.	23.	23.A	24.	10.A	10.A	3.A	95.A	29.A	28.	35.A	91.	22.A	6.

TSP/PM10 NITRATE (nanomoles/m3)

Prd	BD1	BD3	CF1	CF3	CF2	GF1	GF2	GF3	KD1	KD2	MF1
12	.	.	35.	.	36.	40.	48.	42.	65.	.	.
13	.	.	55.	.	55.	53.	77.	67.	96.	.	63.
14	.	.	78.	.	77.	97.	96.	82.	108.	.	87.
15	.	.	51.	.	51.	54.	62.	62.	61.	.	65.
21	.	.	83.	.	79.	.	.	.	87.	.	.
22	.	.	136.	.	145.	121.	.	125.	238.	.	144.
23	.	.	140.	.	164.	.	.	139.	245.	.	.
24	.	.	227.	.	224.	212.	.	245.	363.	.	255.
25	.	.	191.	.	196.	209.	.	214.	242.	.	200.
31	.	.	140.	.	140.	155.	.	152.	165.	.	145.
32	.	.	172.	.	.	153.	.	165.	264.	.	.
33	.	.	125.	.	.	131.	.	135.	198.	.	.
34	.	.	161.	.	.	183.	.	190.	253.	.	.
35	.	.	95.	.	.	109.	.	100.	142.	.	.
41	.	.	178.	.	.	256.	.	234.	225.	.	.
42	.	.	.	.	.	495.	.	522.	636.	.	.
43	.	.	193.	.	.	191.	.	179.	385.	.	.
44	.	.	90.	.	.	111.	.	109.	133.	.	.
45	.	.	99.	.	.	103.	.	104.	121.	.	.
51	.	.	99.	.	.	124.	.	124.	131.	.	.
52	.	.	.	.	.	510.	.	531.	679.	.	.
53	.	.	116.	.	.	105.	.	102.	297.	.	.
54	.	.	182.	.	.	220.	.	208.	224.	.	.
55	.	.	219.	.	.	276.	.	257.	261.	.	.
61	.	.	235.	.	.	299.	.	326.	289.	.	.
62	.	.	.	.	.	721.	.	705.	712.	.	.
63	.	.	229.	.	.	237.	.	229.	406.	.	.
64	.	.	182.	.	.	192.	.	193.	192.	.	.
65	.	.	188.	.	.	256.	.	252.	238.	.	.
71	.	.	.	.	.	399.	.	401.	330.	.	.
72	.	.	.	423.	.	624.	.	633.	622.	.	.
73	.	.	276.	281.	.	284.	.	258.	413.	.	.
74	.	.	226.	231.	.	292.	.	226.	237.	.	.
75	.	.	147.	.	.	152.	.	141.	135.	.	.
81	.	.	117.	.	.	129.	.	124.	118.	.	.
82	.	.	91.	.	.	101.	.	95.	98.	.	94.
83	.	.	83.	.	.	92.	.	87.	101.	.	.
84	.	.	47.	.	.	50.	.	46.	54.	.	55.
85	.	.	59.	.	.	58.	.	58.	62.	.	150.
91	.	.	.	62.	.	68.	.	67.	61.	.	74.
16	68.	65.	56.A	.	56.A	63.A	74.A	64.A	89.A	.	.
17	79.	72.	70.A	.	68.A	.	.	.	77.A	.	.
26	226.	219.	168.A	.	178.A	.	.	169.A	282.A	.	.
27	170.	162.	160.A	.	162.A	177.A	.	177.A	196.A	.	167.A
36	185.	190.	153.A	.	.	155.A	.	163.A	238.A	.	.
37	167.	160.	145.A	.	.	197.A	.	181.A	192.A	.	.
46	329.	360.	210.A	.	.	266.A	.	270.A	385.A	.	.
47	108.	109.	99.A	.	.	116.A	.	116.A	127.A	.	.
56	345.	365.	222.A	.	.	278.A	.	280.A	400.A	.	.
57	234.	210.	229.A	.	.	290.A	.	298.A	278.A	.	.
66	353.	364.	274.A	.	.	383.A	.	376.A	437.A	.	.
67	275.	269.	263.A	.	.	342.A	.	341.A	293.A	.	.
76	342.	349.	315.A	312.A	.	400.A	.	372.A	424.A	.	.
77	124.	114.	129.A	.	.	138.A	.	131.A	125.A	.	.
86	82.	82.	74.A	.	.	81.A	.	76.A	84.A	.	.
87	64.	67.	.	.	.	64.A	.	64.A	61.A	.	105.A
18	73.A	68.A	63.A	.	61.A	.	.	.	84.A	88.	.
28	200.A	193.A	164.A	.	171.A	.	171.	173.A	243.A	278.	.
38	176.A	176.A	149.A	.	.	174.A	153.	171.A	217.A	259.	.
48	229.A	246.A	160.A	.	.	197.A	150.	200.A	268.A	314.	.
58	295.A	294.A	225.A	.	.	283.A	224.	289.A	344.A	358.	.
68	318.A	321.A	269.A	.	.	364.A	365.	360.A	372.A	401.	.
78	243.A	242.A	230.A	.	.	281.A	248.	263.A	288.A	300.	.
88	74.A	75.A	.	.	.	73.A	82.	70.A	74.A	70.	.

FINE SULFATE (nanmomoles/m3)

	BD1	BD3	DE1	DE2	FD1	GF1	GF2	GF3	HE1	HE2	IA1	IA2	JF1	KD1	KD2	NF1	NF2	QA1
12	.	.	.	.	13.	18.	18.	17.	.	.	11.	12.	.	16.	.	.	.	3.
13	.	.	.	.	15.	28.	26.	25.	.	.	19.	19.	.	20.	.	.	.	15.
14	.	.	.	.	21.	17.	23.	22.	.	.	19.	16.	.	21.	.	.	.	3.
15	.	.	.	.	13.	27.	15.	15.	.	.	13.	10.	.	12.	.	.	.	8.
21	.	.	.	.	17.	0.	0.	0.	.	.	15.	15.	.	15.	.	.	.	0.
22	.	.	.	.	21.	23.	.	21.	.	.	17.	15.	.	17.	.	.	.	3.
23	.	.	.	.	12.	13.	.	13.	.	.	12.	10.	.	11.	.	.	.	9.
24	.	.	.	.	25.	28.	.	28.	.	.	22.	21.	.	26.	.	.	.	3.
25	.	.	.	.	14.	33.	.	31.	.	.	22.	26.	.	26.	.	.	.	21.
31	.	.	.	.	21.	18.	.	19.	.	.	15.	15.	.	15.	.	.	.	7.
32	.	.	.	.	17.	18.	.	26.	.	.	12.	11.	.	12.	.	.	.	13.
33	.	.	.	.	20.	20.	.	23.	.	.	17.	14.	.	10.	.	.	.	15.
34	.	.	.	.	42.	50.	.	31.	.	.	40.	0.	.	42.	.	.	.	33.
35	.	.	.	.	21.	34.	.	36.	.	.	29.	0.	.	33.	.	.	.	23.
41	.	.	.	.	30.	39.	.	34.	.	.	27.	0.	.	30.	.	.	.	10.
42	.	.	.	.	42.	35.	.	35.	.	.	32.	.	.	28.	.	.	.	10.
43	.	.	.	.	26.	35.	.	36.	.	.	30.	.	.	31.	.	.	.	23.
44	.	.	.	.	38.	39.	.	38.	.	.	24.	.	.	35.	.	.	.	3.
45	.	.	.	.	22.	32.	.	33.	.	.	15.	.	.	24.	.	.	.	22.
51	.	.	.	.	25.	35.	.	33.	.	.	28.	.	.	25.	.	.	.	23.
52	.	.	.	.	29.	41.	.	42.	.	.	34.	.	.	41.	.	.	.	25.
53	.	.	.	.	27.	35.	.	36.	.	.	30.	.	.	35.	.	.	.	27.
54	.	.	.	.	19.	39.	.	39.	.	.	32.	.	.	38.	.	.	.	7.
55	.	.	.	.	26.	32.	.	32.	.	.	24.	.	.	32.	.	.	.	30.
61	.	.	.	.	32.	42.	.	42.	.	.	32.	.	.	44.	.	.	.	13.
62	.	.	.	.	75.	89.	.	0.	.	.	72.	0.	.	76.	.	.	.	22.
63	.	.	.	.	29.	45.	.	0.	.	.	29.	0.	.	45.	.	.	.	20.
64	.	.	.	.	27.	30.	.	0.	.	.	0.	19.	.	28.	.	.	.	18.
65	.	.	.	.	31.	41.	.	0.	.	.	0.	0.	.	40.	.	.	.	18.
71	.	.	.	.	57.	76.	.	0.	.	.	0.	0.	.	70.	.	.	.	17.
72	.	.	.	.	56.	84.	.	0.	.	.	.	62.	.	75.	.	.	.	54.
73	.	.	.	.	38.	48.	.	0.	.	.	.	40.	.	45.	.	.	.	33.
74	.	.	.	.	41.	47.	.	0.	.	.	.	42.	.	42.	.	.	.	31.
75	.	.	.	.	44.	30.	.	0.	.	.	.	21.	.	29.	.	.	.	18.
81	.	.	.	.	40.	27.	.	0.	.	.	.	17.	.	24.	.	.	.	18.
82	.	.	.	.	32.	21.	.	0.	.	.	15.	18.	.	21.	.	.	.	12.
83	.	.	.	.	4.	20.	.	0.	.	.	14.	14.	.	22.	.	.	.	11.
84	.	.	.	.	3.	12.	.	0.	.	.	8.	8.	.	13.	.	.	.	8.
85	.	.	.	.	8.	15.	.	0.	.	.	12.	12.	.	17.	.	.	.	10.
91	.	.	.	.	16.	17.	.	0.	.	.	14.	13.	.	18.	.	.	.	12.
16	17.	18.	22.	.	16.	21.A	22.	22.A	22.	.	16.A	15.A	0.	19.A	.	4.	.	7.A
17	16.	15.	7.	.	15.	0.	0.	0.	17.	.	14.A	13.A	0.	14.A	.	40.	.	.
26	16.	16.	.	.	19.	21.A	.	21.A	23.	.	17.A	15.A	0.	18.A	.	45.	.	5.A
27	19.	19.	.	.	18.	24.A	.	24.A	22.	.	18.A	19.A	56.	19.A	.	72.	.	13.A
36	23.	24.	0.	.	26.	29.A	.	27.A	27.	.	23.A	0.	0.	22.A	.	51.	.	20.A
37	29.	29.	36.	.	26.	37.A	.	34.A	34.	.	28.A	28.	56.	31.A	.	65.	.	15.A
46	28.	29.	27.	.	35.	36.A	.	36.A	37.	.	29.A	31.	53.	31.A	.	56.	.	12.A
47	27.	26.	23.	.	24.	34.A	.	33.A	32.	.	23.A	28.	54.	25.A	.	44.	.	23.A
56	28.	30.	37.	.	25.	38.A	.	39.A	39.	.	32.A	34.	55.	38.A	.	55.	.	19.A
57	33.	30.	23.	.	30.	38.A	.	38.A	33.	.	29.A	31.	56.	39.A	.	77.	.	20.A
66	41.	45.	40.	.	44.	55.A	.	0.	46.	.	.	.	57.	50.A	.	0.	.	20.A
67	51.	47.	28.	.	47.	62.A	.	0.	51.	.	.	45.	62.	58.A	.	146.	.	18.A
76	44.	45.	58.	.	45.	60.A	.	0.	52.	.	47.	48.A	59.	54.A	.	115.	.	40.A
77	23.	22.	23.	.	42.	28.A	.	0.	25.	.	23.	19.A	.	26.A	.	77.	.	18.A
86	15.	14.	21.	.	13.	18.A	.	0.	15.	.	12.A	13.A	.	19.A	.	26.	.	10.A
87	14.	14.	13.	.	13.	16.A	.	0.	15.	.	13.A	12.A	.	18.A	.	31.	.	11.A
18	17.A	17.	15.A	.	16.	0.	0.	0.	19.A	19.	15.A	14.A	0.A	17.A	17.	20.A	47.	.
28	18.A	17.	.	.	19.	23.A	36.	22.A	22.A	20.	17.A	17.A	26.A	19.A	19.	57.A	.	8.A
38	26.A	26.	.	32.	26.	33.A	50.	30.A	31.A	31.	25.A	0.	26.A	26.A	32.	57.A	.	18.A
48	27.A	28.	25.A	0.	30.	35.A	58.	35.A	34.A	35.	26.A	30.A	53.A	29.A	33.	51.A	0.	17.A
58	30.A	30.	31.A	41.	27.	38.A	53.	39.A	36.A	34.	31.A	32.A	55.A	39.A	39.	65.A	0.	20.A
68	45.A	46.	34.A	43.	45.	58.A	86.	0.	49.A	52.	.	.	60.A	53.A	55.	0.	0.	19.A
78	34.A	35.	41.A	43.	44.	45.A	61.	0.	41.A	41.	36.A	35.A	0.	41.A	41.	98.A	.	30.A
88	14.A	14.	17.A	15.	13.	17.A	26.	0.	15.A	17.	13.A	13.A	.	18.A	18.	28.A	53.	11.A

TSP/PM10 SULFATE (nanomoles/m3)

	BD1	BD3	CF1	CF2	FO1	FO2	FO3	GF1	GF2	GF3	KD1	KD2
12	.	.	19.	21.	.	.	.	27.	24.	26.	9.	.
13	.	.	28.	30.	.	.	.	30.	40.	35.	8.	.
14	.	.	29.	30.	.	.	.	33.	32.	33.	4.	.
15	.	.	20.	20.	.	.	.	21.	26.	26.	10.	.
21	.	.	32.	30.	.	.	.	0.	0.	0.	22.	.
22	.	.	26.	26.	.	.	.	24.	.	26.	6.	.
23	.	.	17.	18.	.	.	.	0.	.	18.	2.	.
24	.	.	35.	35.	.	.	.	34.	.	39.	1.	.
25	.	.	42.	43.	.	.	.	44.	.	47.	9.	.
31	.	.	48.	49.	.	.	.	56.	.	54.	35.	.
32	.	.	23.	.	.	.	.	20.	.	22.	5.	.
33	.	.	26.	.	.	.	.	26.	.	28.	1.	.
34	.	.	59.	.	.	.	.	60.	.	61.	8.	.
35	.	.	42.	.	.	.	.	42.	.	40.	6.	.
41	.	.	56.	.	.	.	.	88.	.	64.	47.	.
42	.	.	42.	.	.	.	.	42.	.	33.	7.	.
43	.	.	43.	.	.	.	.	42.	.	42.	4.	.
44	.	.	42.	.	.	.	.	43.	.	42.	0.	.
45	.	.	47.	.	.	.	.	45.	.	47.	11.	.
51	.	.	49.	.	.	.	.	66.	.	59.	27.	.
52	.	.	48.	.	.	.	.	50.	.	49.	6.	.
53	.	.	41.	.	.	.	.	39.	.	46.	6.	.
54	.	.	48.	.	.	.	.	51.	.	50.	9.	.
55	.	.	45.	.	.	.	.	48.	.	47.	10.	.
61	.	.	58.	.	.	.	.	70.	.	70.	25.	.
62	.	.	107.	.	.	.	.	121.	.	117.	36.	.
63	.	.	55.	.	.	.	.	57.	.	56.	10.	.
64	.	.	43.	.	.	.	.	42.	.	45.	10.	.
65	.	.	59.	.	.	.	.	63.	.	60.	21.	.
71	.	.	93.	.	.	.	.	107.	.	99.	27.	.
72	.	.	92.	.	.	.	.	103.	.	107.	10.	.
73	.	.	54.	.	.	.	.	62.	.	59.	3.	.
74	.	.	58.	.	.	.	.	67.	.	60.	10.	.
75	.	.	42.	.	.	.	.	43.	.	42.	7.	.
81	.	.	36.	.	.	.	.	42.	.	39.	10.	.
82	.	.	29.	.	.	.	.	32.	.	30.	10.	.
83	.	.	29.	.	.	.	.	35.	.	30.	12.	.
84	.	.	19.	.	.	.	.	20.	.	18.	16.	.
85	.	.	23.	.	.	.	.	23.	.	21.	10.	.
91	.	.	24.	.	.	.	.	25.	.	25.	6.	.
16	.	.	25.A	27.A	.	.	.	30.A	32.A	32.A	7.A	.
17	.	.	27.A	26.A	.	.	.	0.	0.	0.	17.A	.
26	.	.	26.A	26.A	.	.	.	0.	.	28.A	3.A	.
27	.	.	46.A	47.A	.	.	.	51.A	.	51.A	25.A	.
36	.	.	36.A	.	.	.	.	35.A	.	37.A	5.A	.
37	.	.	50.A	.	.	.	.	70.A	.	54.A	30.A	.
46	.	.	42.A	.	.	.	.	42.A	.	39.A	3.A	.
47	.	.	48.A	.	.	.	.	58.A	.	54.A	21.A	.
56	.	.	46.A	.	.	.	.	46.A	.	48.A	7.A	.
57	.	.	53.A	.	.	.	.	61.A	.	61.A	19.A	.
66	.	.	68.A	.	.	.	.	74.A	.	73.A	19.A	.
67	.	.	79.A	.	.	.	.	89.A	.	84.A	24.A	.
76	.	.	68.A	.	.	.	.	77.A	.	75.A	8.A	.
77	.	.	38.A	.	.	.	.	43.A	.	40.A	8.A	.
86	.	.	26.A	.	.	.	.	29.A	.	26.A	12.A	.
87	.	.	24.A	.	.	.	.	24.A	.	23.A	8.A	.
18	.	.	26.A	27.A	50.	35.	34.	0.	0.	0.	11.A	9.
28	.	.	35.A	36.A	64.	63.	57.	0.	57.	39.A	13.A	17.
38	.	.	43.A	.	83.	78.	70.	51.A	79.	45.A	16.A	20.
48	.	.	45.A	.	55.	60.	54.	49.A	79.	46.A	11.A	16.
58	.	.	49.A	.	60.	61.	51.	53.A	84.	54.A	12.A	13.
68	.	.	73.A	.	57.	53.	47.	81.A	121.	78.A	21.A	18.
78	.	.	55.A	.	51.	50.	46.	62.A	86.	59.A	8.A	16.
88	.	.	25.A	.	47.	37.	33.	27.A	39.	25.A	10.A	9.

AMMONIA (nanomoles/m3)

	AC1	AF1	AO1	CF1	CF2	DE1	DE2	GF1	GF3	HE1	HE2	IA.	IA2	QA1	RC1	TC1
12	.	149.	.	107.	111.	.	.	159.	238.	.	.	.	.	122.	97.	18.
13	.	186.	.	60.	59.	.	.	248.	198.	.	.	.	.	85.	81.	21.
14	.	174.	.	63.	66.	.	.	109.	134.	.	.	.	.	117.	102.	20.
15	.	177.	.	132.	119.	.	.	191.	124.	.	.	.	.	121.	0.	54.
21	.	178.	.	125.	109.	.	.	.	.	.	.	.	.	0.	0.	29.
22	.	308.	.	177.	164.	.	.	342.	204.	.	.	.	.	173.	205.	37.
23	.	1428.	.	1016.	1189.	.	.	.	1174.	.	.	.	.	912.	1290.	179.
24	.	422.	.	123.	184.	.	.	278.	214	.	.	.	.	121.	238.	49.
25	.	291.	.	163.	180.	.	.	163.	178.	.	.	.	.	159.	193.	43.
31	.	236.	.	149.	152.	.	.	142.	149.	.	.	.	.	135.	125.	30.
32	.	678.	.	502.	0.	.	.	564.	487.	.	.	.	.	426.	567.	53.
33	.	527.	.	325.	.	.	.	319.	265.	.	.	.	.	278.	325.	19.
34	.	389.	.	65.	.	.	.	212.	202.	.	.	.	.	184.	0.	19.
35	.	239.	.	91.	.	.	.	95.	113.	.	.	.	.	121.	0.	24.
41	.	197.	.	109.	.	.	.	121.	107.	.	.	.	.	118.	0.	25.
42	0.	255.	.	130.	.	.	.	284.	178.	.	.	.	.	131.	162.	17.
43	104.	299.	.	117.	.	.	.	238.	203.	.	.	.	.	143.	172.	11.
44	136.	252.	.	62.	.	.	.	176.	126.	.	.	.	.	161.	190.	4.
45	213.	194.	.	131.	.	.	.	182.	.	.	.	.	.	142.	155.	4.
51	0.	154.	.	114.	.	.	.	108.	92.	.	.	.	.	117.	103.	4.
52	0.	369.	.	231.	.	.	.	409.	232.	.	.	.	.	219.	245.	0.
53	110.	431.	.	192.	.	.	.	379.	371.	.	.	.	.	256.	233.	0.
54	154.	204.	.	68.	.	.	.	162.	109.	.	.	.	.	181.	152.	0.
55	290.	220.	.	153.	.	.	.	119.	88.	.	.	.	.	117.	135.	0.
61	0.	128.	.	113.	.	.	.	[illegible]	106.	.	.	.	.	143.	85.	0.
62	320.	961.	.	957.	.	.	.	1070.	.	.	.	.	.	747.	1108.	0.
63	335.	399.	.	308.	.	.	.	530.	493.	.	.	.	.	353.	315.	0.
64	138.	152.	.	114.	.	.	.	133.	.	.	.	.	.	182.	135.	0.
65	165.	112.	.	140.	.	.	.	109.	114.	.	.	.	.	159.	0.	0.
71	0.	167.	.	108.	.	.	.	149.	142.	.	.	.	.	136.	150.	0.
72	792.	0.	.	1048.	.	.	.	1350.	1280.	.	.	..	462.	475.	1185.	.
73	467.	402.	.	266.	.	.	.	534.	498.	.	.	..	1101.	360.	335.	.
74	0.	150.	.	63.	.	.	.	185.	169.	.	.	..	194.	128.	105.	.
75	172.	145.	.	154.	.	.	.	140.	175.	.	.	..	187.	129.	128.	.
81	0.	111.	.	111.	.	.	.	127.	156.	.	.	.	142.	167.	102.	.
82	190.	116.	.	84.	.	.	.	102.	134.	.	.	111.	149.	88.	92.	.
83	138.	73.	.	71.	.	.	.	99.	116.	.	.	78.	105.	80.	84.	.
84	0.	99.	.	137.	.	.	.	163.	161.	.	.	152.	14.	99.	143.	.
85	179.	142.	.	128.	.	.	.	74.	148.	.	.	134.	102.	93.	110.	.
91	126.	59.	.	97.	.	.	.	88.	134.	.	.	82.	73.	70.	70.	.
16	.	170.A	111.	77.A	79.A	72.	.	172.A	186.A	51.	.	.	.	108.A	93.A	20.
17	.	178.A	78.	128.A	113.A	102.	.	.	.	77.	.	.	.	0.	0.	38.
26	.	719.A	572.	439.A	512.A	0.	.	.	531.A	383.	.	.	.	402.A	578.A	101.
27	.	258.A	133.	95.A	163.A	0.	.	150.A	161.A	163.	.	.	.	145.A	152.A	33.
36	.	531.A	344.	297.A	.	0.	.	372.A	318.A	235.	.	.	.	296.A	0.	31.
37	.	214.A	111.	102.A	.	160.	.	111.A	109.A	107.	.	.	.	119.A	0.	24.
46	0.	269.A	167.	103.A	.	245.	.	233.A	169.A	107.	.	.	.	145.A	175.A	10.
47	0.	170.A	78.	121.A	.	110.	.	138.A	.	104.	.	.	.	127.A	124.A	4.
56	0.	335.A	183.	164.A	.	221.	.	317.A	237.A	163.	.	.	.	219.A	210.A	0.
57	0.	165.A	0.	129.A	.	131.	.	.	99.A	109.	.	.	.	133.A	105.A	0.
66	264.A	504.A	400.	485.A	.	433.	.	578.A	.	419.	.	.	.	427.A	519.A	0.
67	0.	145.A	117.	121.A	.	167.	.	133.A	131.A	110.	.	.	.	145.A	0.	0.
76	0.	0.	461.	475.A	.	433.	.	690.A	649.A	399.	.	568.	585.A	321.A	542.A	.
77	0.	124.A	50.	128.A	.	174.	.	132.A	164.A	102.	.	163.	16. A	152.A	112.A	.
86	0.	96.A	0.	97.A	.	95.	.	121.A	137.A	73.	.	114.A	132.A	89.A	106.A	.
87	147.A	92.A	178.	109.A	.	113.	.	82.A	140.A	77.	.	103.A	84.A	79.A	86.A	.
18	0.	173.A	96.A	100.A	94.A	86.A	.	178.A	.	63.A	78.	.	.	0.	0.	27.
28	0.	510.A	373.A	282.A	354.A	.	.	.	362.A	284.A	268.	.	.	285.A	384.A	69.
38	0.	387.A	238.A	209.A	.	.	270.	253.A	223.A	177.A	169.	.	.	216.A	0.	27.
48	0.	224.A	126.A	111.A	.	184.A	.	189.A	.	106.A	93.	.	.	137.A	152.A	7.
58	0.	258.A	0.	148.A	.	181.A	133.	.	174.A	139.A	127.	.	.	180.A	162.A	0.
68	0.	341.A	271.A	320.A	.	312.A	236.	376.A	.	278.A	233.	.	.	299.A	0.	0.
78	0.	0.	274.A	318.A	.	315.A	313.	436.A	428.A	264.A	255.	384.A	392.A	244.A	347.A	0.
88	0.	94.A	0.	103.A	.	104.A	107.	104.A	138.A	75.A	69.	109.A	11. A	85.A	97.A	0.

AMMONIUM ION (nanomoles/m3)

	AC1	AF1	AO1	BD1	BD3	CF1	CF2	DE1	DE2	GF1	GF3	HE1	HE2	IA1	IA2	TC1
12	.	31.	.	.	.	31.	28.	.	.	43.	59.	.	.	23.	19.	18.
13	.	54.	.	.	.	60.	53.	.	.	72.	85.	.	.	39.	34.	12.
14	.	57.	.	.	.	67.	59.	.	.	79.	74.	.	.	36.	18.	13.
15	.	35.	.	.	.	39.	32.	.	.	73.	46.	.	.	19.	11.	12.
21	.	71.	.	.	.	71.	63.	.	.	.	.	.	.	25.	26.	4.
22	.	130.	.	.	.	98.	96.	.	.	112.	102.	.	.	26.	25.	9.
23	.	143.	.	.	.	116.	115.	.	.	136.	114.	.	.	29.	26.	24.
24	.	203.	.	.	.	195.	170.	.	.	238.	243.	.	.	41.	39.	13.
25	.	167.	.	.	.	165.	154.	.	.	208.	209.	.	.	59.	53.	21.
31	.	98.	.	.	.	101.	91.	.	.	109.	109.	.	.	33.	34.	13.
32	.	146.	.	.	.	127.	0.	.	.	107.	121.	.	.	27.	20.	27.
33	.	103.	.	.	.	114.	.	.	.	130.	117.	.	.	28.	28.	13.
34	.	173.	.	.	.	166.	.	.	.	194.	214.	.	.	86.	0.	17.
35	.	114.	.	.	.	111.	.	.	.	142.	139.	.	.	62.	0.	10.
41	.	192.	.	.	.	100.	.	.	.	208.	222.	.	.	67.	0.	19.
42	0.	199.	.	.	.	295.	.	.	.	543.	582.	.	.	61.	.	52.
43	0.	214.	.	.	.	186.	.	.	.	214.	243.	.	.	55.	.	42.
44	0.	101.	.	.	.	106.	.	.	.	132.	149.	.	.	36.	.	8.
45	0.	91.	.	.	.	101.	.	.	.	131.	109.	.	.	28.	.	2.
51	0.	108.	.	.	.	99.	.	.	.	149.	147.	.	.	56.	.	1.
52	0.	207.	.	.	.	313.	.	.	.	571.	581.	.	.	100.	.	0.
53	0.	104.	.	.	.	118.	.	.	.	162.	122.	.	.	58.	.	3.
54	0.	112.	.	.	.	126.	.	.	.	167.	197.	.	.	68.	.	10.
55	0.	157.	.	.	.	152.	.	.	.	207.	220.	.	.	37.	.	29.
61	0.	208.	.	.	.	181.	.	.	.	286.	303.	.	.	48.	.	18.
62	0.	651.	.	.	.	626.	.	.	.	788.	944.	.	.	194.	0.	22.
63	0.	197.	.	.	.	166.	.	.	.	228.	249.	.	.	57.	0.	22.
64	0.	88.	.	.	.	97.	.	.	.	114.	129.	.	.	0.	19.	9.
65	0.	202.	.	.	.	197.	.	.	.	238.	282.	.	.	0.	0.	10.
71	0.	346.	.	.	.	357.	.	.	.	436.	455.	.	.	0.	0.	6.
72	0.	161.	.	.	.	631.	.	.	.	705.	706.	.	.	.	302.	.
73	0.	251.	.	.	.	217.	.	.	.	288.	289.	.	.	.	77.	.
74	0.	132.	.	.	.	150.	.	.	.	217.	228.	.	.	.	80.	.
75	0.	111.	.	.	.	120.	.	.	.	149.	165.	.	.	.	41.	.
81	0.	94.	.	.	.	95.	.	.	.	127.	152.	.	.	0.	38.	.
82	0.	74.	.	.	.	78.	.	.	.	94.	109.	.	.	30.	44.	.
83	0.	56.	.	.	.	65.	.	.	.	87.	85.	.	.	24.	19.	.
84	0.	27.	.	.	.	27.	.	.	.	68.	46.	.	.	18.	12.	.
85	0.	51.	.	.	.	54.	.	.	.	122.	69.	.	.	24.	31.	.
91	0.	46.	.	.	.	48.	.	.	.	71.	53.	.	.	19.	23.	.
16	.	47.A	0.	41.	45.	53.A	47.	.	.	65.A	.	52.	.	33.A	24.A	15.
17	.	56.A	0.	60.	55.	58.A	51.	.	.	.	.	68.	.	23.A	20.A	7.
26	.	159.A	0.	83.	89.	136.A	127.	.	.	162.A	153.A	163.	.	32.A	30.A	17.
27	.	126.A	0.	133.	123.	127.A	116.	.	.	149.A	149.A	120.	.	43.A	41.A	15.
36	.	141.A	0.	80.	94.	136.A	.	.	.	144.A	151.A	85.	.	47.A	0.	19.
37	.	161.A	0.	164.	158.	104.A	.	53.	.	182.A	189.A	144.	.	65.A	65.	16.
46	0.	172.A	0.	99.	98.	196.A	.	41.	.	296.A	324.A	136.	.	51.A	63.	33.
47	0.	101.A	0.	99.	88.	100.A	.	42.	.	142.A	132.A	105.	.	45.A	62.	1.
56	0.	141.A	0.	75.	76.	186.A	.	38.	.	300.A	300.A	126.	.	75.A	67.	7.
57	0.	188.A	0.	136.	138.	169.A	.	0.	.	254.A	270.A	131.	.	43.A	59.	22.
66	0.A	312.A	0.	114.	114.	296.A	.	40.	.	377.A	441.A	305.	.	0.	0.	18.
67	0.	288.A	0.	228.	234.	293.A	.	41.	.	357.A	386.A	213.	.	0.	131.	8.
76	0.	181.A	0.	131.	135.	333.A	.	0.	.	403.A	408.A	359.	.	115.	153.A	.
77	0.	101.A	0.	66.	67.	105.A	.	0.	.	136.A	157.A	104.	.	54.	39.A	.
86	0.	53.A	0.	25.	25.	57.A	.	0.	.	83.A	80.A	44.	.	24.A	25.A	.
87	0.A	48.A	0.	53.	48.	50.A	.	0.	.	91.A	59.A	54.	.	21.A	26.A	.
18	0.	51.A	0.A	49.A	50.A	55.A	49.	.	.	67.A	.	59.A	48.	28.A	22.A	12.
28	0.	144.A	0.A	106.A	105.A	132.A	122.	.	.	156.A	151.A	143.A	104.	37.A	35.A	16.
38	0.	150.A	0.A	118.A	123.A	122.A	0.	.	22.	161.A	168.A	112.A	112.	55.A	0.	17.
48	0.	139.A	0.A	99.A	94.A	152.A	0.	50.A	0.	226.A	463.A	122.A	109.	48.A	62.A	16.
58	0.	162.A	0.	103.A	104.A	178.A	0.	70.A	131.	279.A	286.A	129.A	128.	61.A	63.A	17.
68	0.	301.A	0.A	166.A	169.A	295.A	0.	41.A	203.	368.A	416.A	264.A	302.	0.	0.	12.
78	0.	145.A	0.A	101.A	104.A	229.A	0.	58.A	44.	281.A	294.A	243.A	250.	87.A	101.A	0.
88	0.	51.A	0.	37.A	36.A	54.A	0.	0.A	15.	87.A	71.A	49.A	58.	22.A	26.A	0.

NITROUS ACID (namomoles/m3)

	EA1	ED1	EF1	IA1	IA2	QA1	RC2	TC1
12	14.	25.	43.	16.	30.	24.	24.	0.
13	3.	3.	13.	19.	29.	0.	24.	91.
14	0.	13.	21.	26.	10.	10.	24.	0.
15	0.	0.	0.	66.	51.	58.	26.	0.
21	0.	0.	0.	99.	98.	0.	32.	0.
22	29.	44.	57.	60.	10.	12.	24.	.
23	1.	12.	17.	20.	23.	6.	24.	.
24	57.	0.	5.	58.	54.	33.	24.	.
25	0.	0.	0.	221.	162.	74.	48.	.
31	0.	0.	0.	80.	91.	15.	34.	.
32	41.	4.	8.	62.	10.	15.	24.	.
33	0.	2.	1.	23.	10.	10.	24.	.
34	48.	5.	7.	60.	0.	15.	24.	.
35	0.	0.	0.	165.	0.	25.	40.	.
41	0.	0.	0.	86.	0.	46.	40.	.
42	81.	2.	7.	87.	.	51.	24.	.
43	38.	0.	0.	51.	.	17.	24.	.
44	44.	1.	1.	41.	.	33.	24.	.
45	0.	0.	0.	156.	.	83.	42.	.
51	0.	0.	0.	95.	.	52.	37.	.
52	147.	3.	5.	70.	.	50.	24.	0.
53	29.	4.	4.	31.	.	31.	24.	86.
54	38.	2.	3.	33.	.	29.	24.	124.
55	0.	0.	0.	136.	.	105.	39.	501.
61	0.	0.	0.	192.	.	116.	66.	420.
62	77.	78.	84.	90.	0.	62.	24.	225.
63	24.	2.	4.	10.	0.	12.	24.	97.
64	28.	2.	4.	0.	10.	91.	24.	127.
65	0.	0.	0.	0.	0.	93.	29.	456.
71	0.	0.	0.	0.	0.	78.	26.	305.
72	45.	8.	11.	.	52.	28.	24.	261.
73	33.	4.	6.	.	34.	15.	24.	198.
74	43.	104.	108.	.	30.	36.	24.	273.
75	0.	0.	0.	.	99.	55.	24.	397.
81	0.	0.	0.	.	89.	75.	26.	412.
82	45.	8.	10.	49.	46.	37.	24.	390.
83	36.	10.	13.	44.	34.	21.	24.	298.
84	43.	19.	18.	47.	44.	37.	24.	226.
85	0.	0.	0.	127.	123.	69.	30.	169.
91	0.	0.	0.	57.	57.	71.	24.	46.
16	0.	14.A	26.	21.A	23.A	12.A	24.	0.
17	46.	31.	52.	86.A	79.A	0.	30.	0.
26	29.A	0.	26.	46.A	29.A	17.A	24.	.
27	143.	1.	5.	137.A	120.A	39.A	40.	.
36	3.	4.A	5.	48.A	0.	13.A	24.	.
37	147.	0.	7.	118.A	130.	38.A	40.	.
46	13.	0.	0.	59.A	10.	34.A	24.	.
47	42.	0.	4.	120.A	105.	64.A	39.	.
56	19.	3.A	4.	45.A	10.	36.A	24.	105.
57	180.	2.	5.	169.A	145.	112.A	55.	451.
66	26.	27.A	30.	0.	0.	55.A	24.	133.
67	122.	1.	4.	0.	49.	84.A	27.	371.
76	127.	39.A	41.	37.	38.A	26.A	24.	242.
77	103.	3.	6.	76.	93.A	67.A	25.	410.
86	31.	12.A	14.	47.A	41.A	32.A	24.	295.
87	81.	7.	9.	85.A	83.A	70.A	26.	95.
18	0.	22.A	37.	50.A	48.A	0.	58.	0.
28	81.A	0.	17.	87.A	70.A	27.A	69.	0.
38	68.A	0.	6.	80.A	0.	24.A	69.	0.
48	27.A	0.	0.	87.A	53.A	48.A	67.	0.
58	92.A	2.A	5.	101.A	71.A	71.A	84.	342.
68	70.A	15.A	18.	0.	0.	68.A	56.	260.
78	116.A	22.A	25.	55.A	63.A	45.A	54.	310.
88	54.A	10.A	11.	64.A	60.A	49.A	55.	198.

NO2 (nanomoles/m3)

	CC1	DE1	DE2	HE1	HE2	IA1	IA2	PC1	RC2	TC1
12	805.	.	.	.	.	.	.	590.	810.	.
13	722.	.	.	.	.	.	.	440.	580.	31.
14	1335.	.	.	.	.	.	.	1105.	1120.	.
15	2023.	.	.	.	.	.	.	1944.	2020.	.
21	1624.	.	.	.	.	.	.	1399.	1680.	.
22	2102.	.	.	.	.	.	.	2301.	1890.	.
23	775.	.	.	.	.	103.	109.	827.	850.	.
24	2421.	.	.	.	.	215.	166.	2279.	1990.	.
25	4756.	.	.	.	.	709.	729.	6268.	4550.	.
31	1117.	.	.	.	.	218.	230.	1463.	1473.	.
32	2425.	.	.	.	.	.	.	2305.	2240.	.
33	1015.	.	.	.	.	.	.	857.	1010.	.
34	1797.	.	.	.	.	.	.	2331.	2160.	.
35	3828.	.	.	.	.	.	.	5448.	4560.	.
41	2282.	.	.	.	.	.	.	2203.	2167.	.
42	3158.	.	.	.	.	.	.	0.	2700.	.
43	1000.	.	.	.	.	.	.	906.	980.	.
44	1346.	.	.	.	.	.	.	1107.	1400.	.
45	3211.	.	.	.	.	.	.	3143.	3190.	.
51	1906.	.	.	.	.	.	.	1530.	1913.	.
52	1677.	.	.	.	.	.	.	1519.	1640.	.
53	425.	.	.	.	.	.	.	199.	420.	242.
54	1188.	.	.	.	.	.	.	714.	1100.	468.
55	2794.	.	.	.	.	.	.	2587.	2610.	1037.
61	1218.	.	.	.	.	.	.	1139.	1520.	565.
62	1861.	.	.	.	.	.	.	1658.	1860.	697.
63	726.	.	.	.	.	.	.	399.	720.	558.
64	.	.	.	.	.	.	.	797.	1200.	795.
65	2369.	.	.	.	.	.	.	1993.	2190.	646.
71	1060.	.	.	.	.	.	.	857.	1160.	354.
72	1184.	.	.	.	.	.	239.	812.	1100.	326.
73	869.	.	.	.	.	.	365.	579.	900.	473.
74	1542.	.	.	.	.	.	468.	1512.	1610.	558.
75	2279.	.	.	.	.	.	898.	2497.	2340.	601.
81	1703.	.	.	.	.	.	967.	1718.	1613.	315.
82	1237.	.	.	.	.	.	.	1376.	1330.	279.
83	1309.	.	.	.	.	.	.	1011.	1230.	201.
84	1658.	.	.	.	.	.	.	1767.	1550.	202.
85	1342.	.	.	.	.	.	.	1384.	1550.	53.
91	793.	.	.	.	.	.	.	192.	887.	10.
16	954.A	458.	.	625.	.	.	.	712.A	837.A	.
17	1784.A	1602.	.	1530.	.	.	.	1617.A	1816.A	.
26	1766.A	.	.	1170.	.	.	.	1802.A	1577.A	.
27	2573.A	.	.	2682.	.	415.A	430.A	3385.A	2704.A	.
36	1746.A	.	.	1377.	.	.	.	1831.A	1803.A	.
37	2901.A	2466.	.	2727.	.	.	.	3501.A	3124.A	.
46	1835.A	1296.	.	1746.	.	.	.	671.A	1693.A	.
47	2428.A	1935.	.	2493.	.	.	.	2176.A	2424.A	.
56	1097.A	634.	.	891.	.	.	.	811.A	1053.A	355.
57	1848.A	1512.	.	1800.	.	.	.	1718.A	1956.A	747.
66	862.A	600.	.	927.	.	.	.	951.A	1260.A	656.
67	1584.A	1224.	.	1368.	.	.	.	1312.A	1572.A	482.
76	1198.A	683.	.	1044.	.	203.	357.A	968.A	1203.A	453.
77	1933.A	1665.	.	1629.	.	928.	939.A	2030.A	1904.A	359.
86	1401.A	1080.	.	1170.	.	.	.	1385.A	1370.A	226.
87	1013.A	675.	.	936.	.	.	.	669.A	1152.A	27.
18	1331.A	978.	.	1035.A	1008.	.	.	1123.A	1282.A	.
28	2133.A	.	.	1857.A	1926.	.	.	2522.A	2089.A	.
38	2271.A	.	1863.	1990.A	2097.	.	.	2590.A	2403.A	.
48	2105.A	1587.A	.	2085.A	2160.	.	.	1355.A	2025.A	.
58	1438.A	1033.A	1152.	1304.A	1350.	.	.	1223.A	1464.A	623.
68	1190.A	884.A	963.	1128.A	1278.	.	.	1115.A	1402.A	563.
78	1532.A	1130.A	1071.	1310.A	1503.	532.A	622.A	1450.A	1522.A	415.
88	1225.A	896.A	972.	1064.A	990.	.	.	1059.A	1271.A	130.

SO2 (nanomoles/m3)

	DE1	DE2	HE1	HE2	IA1	IA2	JF1	JF2	NF1	NF2	QA1
12	.	.	.	.	19.	18.	.	.	.	.	21.
13	.	.	.	.	26.	22.	.	.	.	.	16.
14	.	.	.	.	52.	46.	.	.	.	.	34.
15	.	.	.	.	32.	27.	.	.	.	.	36.
21	.	.	.	.	15.	18.	.	.	.	.	.
22	.	.	.	.	60.	52.	.	.	.	.	80.
23	.	.	.	.	30.	27.	.	.	.	.	45.
24	.	.	.	.	62.	91.	.	.	.	.	115.
25	.	.	.	.	74.	70.	.	.	.	.	75.
31	.	.	.	.	19.	29.	.	.	.	.	11.
32	.	.	.	.	49.	56.	.	.	.	.	51.
33	.	.	.	.	50.	49.	.	.	.	.	62.
34	.	.	.	.	152.	.	.	.	.	.	135.
35	.	.	.	.	81.	.	.	.	.	.	49.
41	.	.	.	.	49.	.	.	.	.	.	41.
42	.	.	.	.	114.	.	.	.	.	.	97.
43	.	.	.	.	95.	.	.	.	.	.	104.
44	.	.	.	.	68.	.	.	.	.	.	80.
45	.	.	.	.	62.	.	.	.	.	.	39.
51	.	.	.	.	38.	.	.	.	.	.	37.
52	.	.	.	.	103.	.	.	.	.	.	97.
53	.	.	.	.	69.	.	.	.	.	.	60.
54	.	.	.	.	91.	.	.	.	.	.	84.
55	.	.	.	.	54.	.	.	.	.	.	69.
61	.	.	.	.	31.	.	.	.	.	.	41.
62	.	.	.	.	38.	.	.	.	.	.	44.
63	.	.	.	.	36.	.	.	.	.	.	55.
64	.	.	.	.	.	12.	.	.	.	.	52.
65	.	.	.	.	.	.	.	.	.	.	70.
71	.	.	.	.	.	.	.	.	.	.	34.
72	.	.	.	.	.	14.	.	.	.	.	24.
73	.	.	.	.	.	17.	.	.	.	.	43.
74	.	.	.	.	.	53.	.	.	.	.	90.
75	.	.	.	.	.	57.	.	.	.	.	61.
81	.	.	.	.	.	34.	.	.	.	.	34.
82	.	.	.	.	63.	64.	.	.	.	.	65.
83	.	.	.	.	27.	26.	.	.	.	.	37.
84	.	.	.	.	34.	29.	.	.	.	.	39.
85	.	.	.	.	14.	13.	.	.	.	.	20.
91	.	.	.	.	10.	9.	.	.	.	.	43.
16	138.	.	72.	.	32.A	29.A	20.	0.	11.	.	24.A
17	96.	.	39.	.	22.A	22.A	69.	109.	23.	.	.
26	.	.	144.	.	51.A	57.A	22.	2.	38.	.	80.A
27	.	.	64.	.	41.A	45.A	26.	74.	33.	.	37.A
36	.	.	233.	.	84.A	.	77.	122.	238.	.	83.A
37	236.	.	89.	.	62.A	64.	41.	166.	12.	.	44.A
46	284.	.	240.	.	92.A	10.	118.	64.	306.	.	94.A
47	97.	.	50.	.	48.A	42.	108.	135.	2.	.	38.A
56	185.	.	131.	.	88.A	80.	26.	31.	285.	.	80.A
57	67.	.	64.	.	40.A	38.	99.	98.	23.	.	52.A
66	77.	.	50.	.	.	.	18.	125.	10.	.	50.A
67	77.	.	56.	.	.	28.	24.	20.	13.	.	48.A
76	88.	.	87.	.	41.	28.A	85.	22.	40.	.	52.A
77	86.	.	60.	.	40.	43.A	28.	139.	33.	.	45.A
86	101.	.	56.	.	41.A	40.A	28.	31.	209.	.	47.A
87	54.	.	34.	.	12.A	11.A	0.	91.	.	.	34.A
18	119.A	.	57.A	41.	27.A	26.A	43.A	50.A	17.A	12.	.
28	.	.	108.A	79.	46.A	52.A	24.A	35.A	36.A	.	60.A
38	.	175.	167.A	107.	74.A	.	60.A	142.A	135.A	.	65.A
48	200.A	.	154.A	120.	72.A	25.A	113.A	96.A	168.A	.	68.A
58	131.A	98.	101.A	113.	66.A	61.A	59.A	61.A	166.A	.	67.A
68	77.A	70.	52.A	68.	.	.	20.A	77.A	11.A	.	49.A
78	87.A	50.	75.A	67.	41.A	35.A	59.A	75.A	37.A	.	49.A
88	79.A	64.	46.A	38.	28.A	27.A	15.A	58.A	.	.	41.A